

UNITED STATES DISTRICT COURT  
DISTRICT OF DELAWARE

-----X

SIEMENS MEDICAL SOLUTIONS USA,  
INC.,

Plaintiff,

Civil Action No. 07-190 (SLR)

v.

REDACTED  
PUBLIC VERSION

SAINT-GOBAIN CERAMICS & PLASTICS,  
INC.,

Defendant.

-----X

**DECLARATION OF FREDERICK L. WHITMER**

Frederick L. Whitmer, of full age, declares as follows:

1. I am a member of Thelen Reid Brown Raysman & Steiner LLP, counsel for Defendant Saint-Gobain Ceramics & Plastics, Inc., ("Saint-Gobain") in the above-captioned action. I have been admitted pro hac vice to appear for Saint-Gobain in this action. I submit this declaration upon personal knowledge, in support of Defendant's Motion in Opposition to Plaintiff's Motion for Summary Judgment on Saint-Gobain's Affirmative Defenses and Motion to Strike Saint-Gobain's Fourth Affirmative Defense.

2. Attached hereto as Exhibit 1 is a true and correct copy of the Non-Infringement Opinion for U.S. Patent No. 4,958,080 by Foley & Lardner LLP dated March 16, 2006.

3. Attached hereto as Exhibit 2 is a true and correct copy of a letter from John Ohman, Esq. to Charanjit Brahma, Esq. dated September 20, 2007.

4. Attached hereto as Exhibit 3 is a true and correct copy of Defendant's Responses and Objections to Plaintiff's First Set of Interrogatories.

5. Attached hereto as Exhibit 4 is a true and correct copy of the Foley & Lardner "freedom to operate" Memorandum dated April 28, 2006.

6. Attached hereto as Exhibit 5 is a true and correct copy of Non-Party Subpoena of Leon Radomsky.

7. Attached hereto as Exhibit 6 is a true and correct copy of a letter from Kenneth Krosin, Esq., Foley & Lardner, to Charanjit Brama, Esq., Kirkland & Ellis dated February 19, 2008.

8. Attached hereto as Exhibit 7 is a true and correct copy of United States Patent No. 4,958,080 Patent Prosecution History, Patent Application Serial No. 07/389502.

9. Attached hereto as Exhibit 8 is a true and correct copy of an article entitled "Boron Nitride, A Neutron Scintillator With Deficiencies," IEEE Trans. Nucl. Sci. N35-31 (2005).

10. Attached hereto as Exhibit 9 is a true and correct copy of a Saint-Gobain internal correspondence dated March 7, 2006.

11. Attached hereto as Exhibit 10 is a true and correct copy of a 2006 IEEE Medical Imaging Conference Presentation by Philips concerning its Gemini Raptor PET/CT system with time-of-flight capability.

12. Attached hereto as Exhibit 11 is a true and correct copy of a letter from Mr. Schmidt, Siemens to Mr. Schouten, Philips Intellectual Properties & Standards dated March 17, 2006.


13. Attached hereto as Exhibit 12 is a true and correct copy of an article entitled "Large Size LYSO Crystals for Future High Energy Physics Experiments," IEEE Trans. Nucl. Sci. NS-52 (2005).

14. Attached hereto as Exhibit 13 is a true and correct copy of pages from the deposition transcript of Dominique Rothan dated April 22, 2008.

15. Attached hereto as Exhibit 14 is a true and correct copy of pages from the deposition transcript of Thomas Field dated April 9, 2008.

I declare under penalty of perjury that the foregoing is true and correct.

Executed on May 30, 2008.

  
\_\_\_\_\_  
FREDERICK L. WHITMER

UNITED STATES DISTRICT COURT  
FOR THE DISTRICT OF DELAWARE

CERTIFICATE OF SERVICE

I hereby certify that on May 30, 2008, I electronically filed the foregoing document with the Clerk of the Court using CM/ECF which will send notification of such filing to the following and which has also been served as noted:

BY E-MAIL AND HAND DELIVERY

Jack B. Blumenfeld  
Maryellen Noreika  
Morris, Nichols, Arsht & Tunnell LLP  
1201 North Market Street  
Wilmington, DE 19899

I hereby certify that on May 30, 2008, the foregoing document was sent to the following non-registered participants in the manner indicated:

BY E-MAIL

Gregg F. LoCascio  
Charanjit Brahma  
Sean M. McEldowney  
Kirkland & Ellis LLP  
655 15<sup>th</sup> Street, N.W.  
Washington, DC 20005-5793

  
\_\_\_\_\_  
Kelly E Parnan (#4395)

UNITED STATES DISTRICT COURT  
FOR THE DISTRICT OF DELAWARE

**CERTIFICATE OF SERVICE**

I hereby certify that on June 30, 2008, I electronically filed the foregoing document with the Clerk of the Court using CM/ECF which will send notification of such filing to the following and which has also been served as noted:

**BY HAND DELIVERY**

Jack B. Blumenfeld  
Maryellen Noreika  
Morris, Nichols, Arsht & Tunnell LLP  
1201 North Market Street  
Wilmington, DE 19899

I hereby certify that on June 30, 2008, the foregoing document was sent to the following non-registered participants in the manner indicated:

**BY FEDERAL EXPRESS**

Gregg F. LoCascio  
Charanjit Brahma  
Sean M. McEldowney  
Kirkland & Ellis LLP  
655 15<sup>th</sup> Street, N.W.  
Washington, DC 20005-5793


  
\_\_\_\_\_  
Kelly E. Farnan (#4395)

EXHIBIT 1  
REDACTED  
IN ITS ENTIRETY

**EXHIBIT 2**

THELEN REID BROWN  
RAYSMAN & STEINER LLP  
ATTORNEYS AT LAW

John C. Ohman  
Partner  
212 603.6784 Direct Dial  
johman@thelen.com

NEW YORK • SAN FRANCISCO • WASHINGTON, DC • LOS ANGELES  
SILICON VALLEY • HARTFORD • NORTHERN NEW JERSEY • SHANGHAI

September 20, 2007

Federal Express

Charanjit Brahma, Esq.  
Kirkland & Ellis LLP  
655 Fifteenth Street, N.W.  
Washington, DC 20005

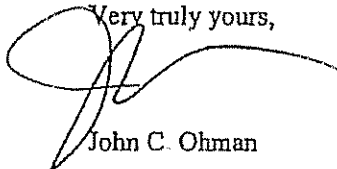
Re: Siemens Medical Solutions USA, Inc. v. Saint-Gobain Ceramics  
& Plastics, Inc. (D. Del. C.A. No. 07-190-SLR)

Dear Charan:

Enclosed are additional documents (Bates Nos. SGCP 001447-002720) that are responsive to Plaintiff's First Set of Requests for Production of Documents. The enclosed documents are being provided to you subject to the agreement memorialized in your August 17, 2007 letter that all documents marked as "confidential," "restricted confidential" "attorney's eyes only" or the like will be treated by the receiving party as "outside counsel's eyes only" until a protective order has been entered by the Court and that, in the event no such order is entered, the documents will be returned.

Defendant reserves the right to supplement its production.

Very truly yours,



John C. Ohman

JCO:esg  
Enclosures

NY #1189242 v1



EXHIBIT 3  
REDACTED  
IN ITS ENTIRETY

EXHIBIT 4  
REDACTED  
IN ITS ENTIRETY

**EXHIBIT 5**

111/57

IN THE UNITED STATES DISTRICT COURT  
FOR THE DISTRICT OF DELAWARE

SIEMENS MEDICAL SOLUTIONS USA, INC.,	)	
	)	
Plaintiff,	)	
	)	
v.	)	C.A. No. 07-190 (SLR)
	)	
SAINT-GOBAIN CERAMICS & PLASTICS, INC.,	)	
	)	
Defendant.	)	

NOTICE OF SUBPOENA

PLEASE TAKE NOTICE that Leon Radomsky, Esquire is being served with the subpoena attached to this notice as Exhibit A.

PLEASE TAKE FURTHER NOTICE that Crystal Photonics, Inc. is being served with the subpoena attached to this notice as Exhibit B.

MORRIS, NICHOLS, ARSHT & TUNNELL LLP

*/s/ Maryellen Noreika*

---

Jack B. Blumenfeld (#1014)  
Maryellen Noreika (#3208)  
1201 North Market Street  
P.O. Box 1347  
Wilmington, DE 19899  
(302) 658-9200  
jblumenfeld@mnat.com  
mnoreika@mnat.com

*Attorneys for Plaintiff  
Siemens Medical Solutions USA, Inc*

*Of Counsel:*

Gregg F. LoCascio  
Charanjit Brahma  
Sean M. McEldowney  
KIRKLAND & ELLIS LLP  
655 15th Street, N.W.  
Washington, D.C. 20005-5793  
(202) 879-5000

February 19, 2008

**CERTIFICATE OF SERVICE**

I, the undersigned, hereby certify that on February 19, 2008, I electronically filed the foregoing with the Clerk of the Court using CM/ECF, which will send notification of such filing(s) to the following:

Kelly E. Farnan, Esquire  
RICHARDS, LAYTON & FINGER, P.A.

I also certify that copies were caused to be served on February 19, 2008 upon the following in the manner indicated:

**BY ELECTRONIC MAIL and HAND DELIVERY**

Kelly E. Farnan, Esquire  
RICHARDS, LAYTON & FINGER, P.A.  
One Rodney Square  
Wilmington, DE 19801

**BY ELECTRONIC MAIL**

Frederick L. Whitmer, Esquire  
THELEN REID BROWN RAYSMAN & STEINER LLP  
875 Third Avenue  
New York, NY 10022

*/s/ Maryellen Norcik*

\_\_\_\_\_  
Maryellen Norcik (#3208)

## EXHIBIT A

AO 88 (Rev. 1/94) Subpoena in a Civil Case

Issued by the  
**UNITED STATES DISTRICT COURT**  
**DISTRICT OF THE DISTRICT OF COLUMBIA**

**SUBPOENA IN A CIVIL CASE**

SIEMENS MEDICAL SOLUTIONS USA,  
 INC.

Case Number: 07-190 (SLR)

v.

United States District Court for the  
 District of Delaware

SAINT-GOBAIN CERAMICS &  
 PLASTICS, INC.

to: Leon Radomsky, Esq.  
 Foley & Lardner LLP  
 3000 K Street, N.W.  
 Washington, DC 20007  
 ATTN: Leon Radomsky

☐ YOU ARE COMMANDED to appear in the United States District Court at the place, date, and time specified below to testify in the above case.

PLACE OF TESTIMONY	COURTROOM
	DATE AND TIME

☒ YOU ARE COMMANDED to appear at the place, date, and time specified below to testify at the taking of a deposition in the above case, to be recorded by stenographic means and/or videotape.

PLACE OF DEPOSITION Kirkland & Ellis LLP 655 Fifteenth St, N.W. Washington, DC 20005 (or by agreement of counsel)	DATE AND TIME March 13, 2008 at 9:00 AM
---	--

☒ YOU ARE COMMANDED to produce and permit inspection and copying of the following documents or objects at the place, date, and time specified below (list documents or objects): **SEE SCHEDULE A**

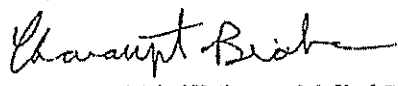
PLACE Attn: Charanjit Brahma Kirkland & Ellis LLP 655 Fifteenth St, N.W. Washington, DC 20005 (or by agreement of counsel)	DATE AND TIME February 29, 2008 at 9:00 AM
---	---



☐ YOU ARE COMMANDED to produce and permit inspection of the following premises at the date and time specified below.

PREMISES	DATE AND TIME
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Any organization not a party to this suit that is subpoenaed for the taking of a deposition shall designate one or more officers, directors, or managing agents, or other persons who consent to testify on its behalf, and may set forth, for each person designated, the matters on which the person will testify. Federal Rules of Civil Procedure, 30(b)(6).

Issuing Officer Signature and Title (Indicate if attorney for Plaintiff or Defendant)	DATE
	
Attorney for Plaintiff Siemens Medical Solutions USA, Inc.	February 15, 2008

Issuing Officer's Name, Address, and Phone Number

Charanjit Brahma, Kirkland & Ellis LLP, 655 Fifteenth Street, N.W., Washington, DC 20005  
(202) 879-5000

(See Rule 45, Federal Rules of Civil Procedure Parts C & D on Reverse)

If action is pending in district other than district of issuance, state district under case number.

AO 88 (Rev. 1/94) Subpoena in a Civil Case

PROOF OF SERVICE

DATE

PLACE

SERVED

SERVED ON (PRINT NAME)

MANNER OF SERVICE

SERVED BY (PRINT NAME)

TITLE

DECLARATION OF SERVER

I declare under penalty of perjury under the laws of the United States of America that the foregoing information contained in the Proof of Service is true and correct.

Executed on

DATE

SIGNATURE OF SERVER

ADDRESS OF SERVER

Rule 45, Federal Rules of Civil Procedure, Parts C & D:

(c) PROTECTION OF PERSONS SUBJECT TO SUBPOENAS.

(1) A party or an attorney responsible for the issuance and service of a subpoena shall take reasonable steps to avoid imposing undue burden or expense on a person subject to that subpoena. The court on behalf of which the subpoena was issued shall enforce this duty and impose upon the party or attorney in breach of this duty an appropriate sanction which may include, but is not limited to, lost earnings and a reasonable attorney's fee.

(2) (A) A person commanded to produce and permit inspection and copying of designated books, papers, documents or tangible things or inspection of premises need not appear in person at the place of production or inspection unless commanded to appear for deposition, hearing or trial.

(B) Subject to paragraph (d)(2) of this rule, a person commanded to produce and permit inspection and copying may, within 14 days after service of the subpoena or before the time specified for compliance if such time is less than 14 days after service, serve upon the party or attorney designated in the subpoena written objection to inspection or copying of any or all of the designated materials or of the premises. If objection is made, the party serving the subpoena shall not be entitled to inspect and copy the materials or inspect the premises except pursuant to an order of the court by which the subpoena was issued. If objection is made, the party serving the subpoena may, upon notice to the person commanded to produce, move at any time for an order to compel the production. Such an order to compel production shall protect any person who is not a party or an officer of a party from significant expense resulting from the inspection and copying commanded.

(3) (A) On timely motion, the court by which a subpoena was issued shall quash or modify the subpoena if it

(i) fails to allow reasonable time for compliance;

(ii) requires a person who is not a party or an officer of a party to travel to a place more than 100 miles from the place where that person resides, is employed or regularly transacts business in person, except that, subject to the provisions of clause (c)(3)(B)(iii) of this rule, such a person may in order to attend

trial be commanded to travel from any such place within the state in which the trial is held, or

(iii) requires disclosure of privileged or other protected matter and no exception or waiver applies, or

(iv) subjects a person to undue burden.

(B) If a subpoena

(i) requires disclosure of a trade secret or other confidential research, development, or commercial information, or

(ii) requires disclosure of an unretained expert's opinion or information not describing specific events or occurrences in dispute and resulting from the expert's study made not at the request of any party, or

(iii) requires a person who is not a party or an officer of a party to incur substantial expense to travel more than 100 miles to attend trial, the court may, to protect a person subject to or affected by the subpoena, quash or modify the subpoena or, if the party in whose behalf the subpoena is issued shows a substantial need for the testimony or material that cannot be otherwise met without undue hardship and assures that the person to whom the subpoena is addressed will be reasonably compensated, the court may order appearance or production only upon specified conditions

(d) DUTIES IN RESPONDING TO SUBPOENA.

(1) A person responding to a subpoena to produce documents shall produce them as they are kept in the usual course of business or shall organize and label them to correspond with the categories in the demand.

(2) When information subject to a subpoena is withheld on a claim that it is privileged or subject to protection as trial preparation materials, the claim shall be made expressly and shall be supported by a description of the nature of the documents, communications, or things not produced that is sufficient to enable the demanding party to contest the claim.

Schedule A

DOCUMENT REQUESTS

1. All Documents and things Concerning any LYSO single crystal scintillator Saint-Gobain sells or offers for sale or has previously sold, offered for sale, or considered offering for sale, including without limitation any information received from or sent to Saint-Gobain regarding such scintillators.
2. All Documents and things Concerning any analysis conducted by or on behalf of Saint-Gobain regarding the '080 Patent, whether Saint-Gobain's products infringe or would infringe the '080 Patent, or the validity or enforceability of the '080 Patent, including without limitation any written or oral opinions of counsel provided to Saint-Gobain, drafts of such opinions, all Documents and things Concerning Saint-Gobain's request for such opinions, all Documents and things Concerning any information about Saint-Gobain's products considered in performing such analyses or preparing such opinions, and all communications regarding such opinions or analyses.
3. All Documents and things Concerning any opinions of counsel, written or otherwise, prepared for and/or provided to Saint-Gobain regarding the '489 Patent or the '420 Patent.
4. Documents sufficient to identify all Foley & Lardner personnel, including without limitation attorneys and patent agents, involved in preparing the March 2006 Opinion, the time spent by each such individual in preparing the March 2006 Opinion, and a description of the tasks performed by each such individual.
5. Documents sufficient to identify all Foley & Lardner personnel, including without limitation attorneys and patent agents, involved in preparing any opinion of counsel for Saint-Gobain regarding either the '489 Patent or the '420 Patent, the time spent by each such individual in preparing such an opinion, and a description of the tasks performed by each such individual.
6. All Documents and things any Foley & Lardner personnel, including without limitation attorneys and patent agents, considered or relied upon in preparing the March 2006 Opinion.
7. All Documents and things Concerning any Foley & Lardner policies regarding the preparation, approval and communication of patent opinions provided to clients, including without limitation the background and qualifications of attorneys permitted to prepare, approve and provide such opinions as well as the review procedure for such opinions prior to their finalization.
8. Documents sufficient to identify the amounts billed to Saint-Gobain or its affiliates by Foley & Lardner for each year from 2003 to the present.
9. Documents sufficient to describe all fees and costs billed by Foley & Lardner to Saint-Gobain for any work Concerning the March 2006 Opinion, the '080 Patent, the '489 Patent, the '420 Patent, LYSO crystals, or LSO crystals, as well as the nature of the work performed.
10. All Documents and things Concerning Saint-Gobain's retention of Foley & Lardner and/or Leon Radomsky to prepare the March 2006 Opinion or to provide advice regarding the '080 Patent, '420 Patent or '489 Patent.
11. A current resume and curriculum vitae for Leon Radomsky.

12. Documents sufficient to identify the title of any patent analyzed in any opinions of counsel provided to a client by Leon Radomsky prior to April 2006 and the dates of the corresponding opinions.
13. Any Documents Concerning this Litigation or Communications Concerning this Litigation, and phone logs or other records of Communications or contact with Saint-Gobain or Philips from January 2006 to the present.

### DEFINITIONS

As used in these requests, the following terms and phrases shall have the following definitions:

1. "Document" as used herein is defined to be synonymous in meaning and equal in scope to the usage of this term in Federal Rule of Civil Procedure 34(a), including, without limitation, electronic or computerized data compilations. A draft or non-identical copy is a separate Document within the meaning of this term.
2. "Concerning" as used herein means relating to, referring to, reflecting, describing, evidencing or constituting.
3. "Communication" as used herein means any transmittal of information (in the form of facts, ideas, inquiries, or otherwise).
4. "Saint-Gobain" as used herein means, individually and collectively, Saint-Gobain Ceramics & Plastics, Inc., and any of its corporate parents, predecessors, successors, and past or present subsidiaries, affiliates, assigns, divisions, departments, officers, directors, principals, agents, representatives and employees.
5. "Philips" as used herein means, individually and collectively, Koninklijke Philips Electronics N.V., and any of its corporate parents, predecessors, successors, and past or present subsidiaries, affiliates, assigns, divisions, departments, officers, directors, principals, agents, representatives and employees.
6. "LSO" as used herein means to the single crystal form of cerium doped lutetium oxyorthosilicate having the general chemical formula of  $Ce_{2x}Lu_{2(1-x)}SiO_5$ .
7. "LYSO" as used herein means to the single crystal form of cerium doped lutetium yttrium oxyorthosilicate having the general chemical formula of  $Ce_{2x}(Lu_{1-y}Y_y)_{2(1-x)}SiO_5$ .
8. "The '080 patent" as used herein means to United States Letters Patent No. 4,958,080.
9. "The '420 patent" as used herein means to United States Letters Patent No. 6,624,420.
10. "The '489 patent" as used herein means to United States Letters Patent No. 6,323,489.
11. "Litigation" as used herein means to Siemens Medical Solutions USA, Inc. v. Saint-Gobain Ceramics & Plastics, Inc., Case Number 07-190, before Judge Robinson in the District of Delaware.
12. "USPTO" as used herein means to the United States Patent and Trademark Office.
13. "Foley & Lardner" as used herein means Foley & Lardner LLP, its attorneys, patent agents, and employees, past and present, located worldwide.

14. "March 2006 Opinion" as used herein means the opinion of counsel provided to Saint-Gobain dated March 16, 2006 entitled "Non-Infringement Opinion for U.S. Patent No. 4,958,080," which has previously been produced by Saint-Gobain bearing the Bates labels SGCP002151-91.

IN THE UNITED STATES DISTRICT COURT  
FOR THE DISTRICT OF DELAWARE

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SIEMENS MEDICAL SOLUTIONS USA, INC.

Plaintiff,

v.

SAINT-GOBAIN CERAMICS & PLASTICS,  
INC.

Defendant.

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Case No. 07-190 (SLR)

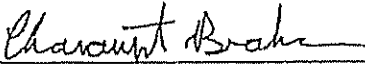
NOTICE OF DEPOSITION OF LEON RADOMSKY PURSUANT TO SUBPOENA

PLEASE TAKE NOTICE that Plaintiff Siemens Medical Solutions USA, Inc. ("Siemens"), by and through its undersigned counsel and pursuant to Federal Rules of Civil Procedure 30 and 45, will take the deposition upon oral examination of Leon Radomsky, Esq., at 9:00 AM on March 13, 2008 at Kirkland & Ellis LLP, 655 Fifteenth Street N.W., Washington, DC 20005, or at such other time or location as may be mutually agreed upon by counsel for Siemens and the deponent.

The deposition will be conducted under oath by an officer authorized to take such testimony and administer oaths. The deposition will be recorded stenographically, and may also be recorded videographically.



Dated: February 15, 2008

By:   
\_\_\_\_\_  
Jack B. Blumenfeld (I.D. No. 1014)  
Maryellen Noreika (I.D. No. 3208)  
MORRIS, NICHOLS, ARSHT & TUNNELL LLP  
1201 North Market Street  
P.O. Box 1347  
Wilmington, DE 19899  
(302) 658-9200  
jblumenfeld@mnat.com

Gregg F. LoCascio  
Charanjit Brahma  
Sean McEldowney  
KIRKLAND & ELLIS LLP  
655 15<sup>th</sup> Street, N.W.  
Washington, DC 20005  
Telephone: (202) 879-5000  
Facsimile: (202) 879-5200

*Attorneys for Plaintiff*  
**SIEMENS MEDICAL SOLUTIONS**  
**USA, INC.**

## EXHIBIT B

AO 88 (Rev. 1/94) Subpoena in a Civil Case

Issued by the  
**UNITED STATES DISTRICT COURT**  
**MIDDLE DISTRICT OF FLORIDA**

**SUBPOENA IN A CIVIL CASE**

**SIEMENS MEDICAL SOLUTIONS USA,  
 INC.**

Case Number: 07-190 (SLR)

v.

United States District Court for the  
 District of Delaware

**SAINT-GOBAIN CERAMICS &  
 PLASTICS, INC.**

TO: Crystal Photonics, Inc.  
 c/o Matthew R. O'Kane  
 Lowndes, Drosdick, Doster, Kantor & Reed, P.A.  
 215 North Eola Drive  
 Orlando, Florida 32802

☐ YOU ARE COMMANDED to appear in the United States District Court at the place, date, and time specified below to testify in the above case.

PLACE OF TESTIMONY	COURTROOM
	DATE AND TIME

☒ YOU ARE COMMANDED to appear at the place, date, and time specified below to testify at the taking of a deposition in the above case, to be recorded by stenographic means and/or videotape, as set forth in the enclosed notice of deposition.

PLACE OF DEPOSITION Hill Ward Henderson 101 East Kennedy Boulevard, Suite 3700 Tampa, Florida 33602 (or by agreement of counsel)	DATE AND TIME March 5, 2008 at 9:00 AM
--	---

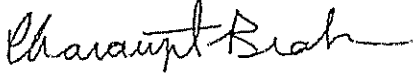
☒ YOU ARE COMMANDED to produce and permit inspection and copying of the following documents or objects at the place, date, and time specified below (list documents or objects): **See attached Schedule A**

PLACE Charanjit Brahma c/o Dennis Waggoner Hill Ward Henderson 101 East Kennedy Boulevard, Suite 3700 Tampa, Florida 33602 (or by agreement of counsel)	DATE AND TIME February 29, 2008 at 9:00 AM
---	---

☐ YOU ARE COMMANDED to produce and permit inspection of the following premises at the date and time specified below.

PREMISES	DATE AND TIME
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Any organization not a party to this suit that is subpoenaed for the taking of a deposition shall designate one or more officers, directors, or managing agents, or other persons who consent to testify on its behalf, and may set forth, for each person designated, the matters on which the person will testify. Federal Rules of Civil Procedure, 30(b)(6).

Issuing Officer Signature and Title (Indicate if attorney for Plaintiff or Defendant)  Attorney for Plaintiff Siemens Medical Solutions USA, Inc.	DATE February 15, 2008
Issuing Officer's Name, Address, and Phone Number Charanjit Brahma, Kirkland & Ellis LLP, 655 Fifteenth Street, N.W., Washington, DC 20005 (202) 879-5000	

(See Rule 45, Federal Rules of Civil Procedure Parts C & D on Reverse)  
 If action is pending in district other than district of issuance, state district under case number.

AO 88 (Rev. 1/99) Subpoena in a Civil Case

PROOF OF SERVICE

DATE

PLACE

SERVED

SERVED ON (PRINT NAME)

MANNER OF SERVICE

SERVED BY (PRINT NAME)

TITLE

DECLARATION OF SERVER

I declare under penalty of perjury under the laws of the United States of America that the foregoing information contained in the Proof of Service is true and correct.

Executed on

DATE

SIGNATURE OF SERVER

ADDRESS OF SERVER

## Rule 45, Federal Rules of Civil Procedure, Parts C &amp; D:

## (c) PROTECTION OF PERSONS SUBJECT TO SUBPOENAS.

(1) A party or an attorney responsible for the issuance and service of a subpoena shall take reasonable steps to avoid imposing undue burden or expense on a person subject to that subpoena. The court on behalf of which the subpoena was issued shall enforce this duty and impose upon the party or attorney in breach of this duty an appropriate sanction which may include, but is not limited to, lost earnings and a reasonable attorney's fee.

(2) (A) A person commanded to produce and permit inspection and copying of designated books, papers, documents or tangible things or inspection of premises need not appear in person at the place of production or inspection unless commanded to appear for deposition, hearing or trial.

(B) Subject to paragraph (d)(2) of this rule, a person commanded to produce and permit inspection and copying may, within 14 days after service of the subpoena or before the time specified for compliance if such time is less than 14 days after service, serve upon the party or attorney designated in the subpoena written objection to inspection or copying of any or all of the designated materials or of the premises. If objection is made, the party serving the subpoena shall not be entitled to inspect and copy the materials or inspect the premises except pursuant to an order of the court by which the subpoena was issued. If objection is made, the party serving the subpoena may, upon notice to the person commanded to produce, move at any time for an order to compel the production. Such an order to compel production shall protect any person who is not a party or an officer of a party from significant expense resulting from the inspection and copying commanded.

(3) (A) On timely motion, the court by which a subpoena was issued shall quash or modify the subpoena if it

- (i) fails to allow reasonable time for compliance;
- (ii) requires a person who is not a party or an officer of a party to travel to a place more than 100 miles from the place where that person resides, is employed or regularly transacts business in person, except that, subject to the provisions of clause (c)(3)(B)(iii) of this rule, such a person may in order to attend

trial be commanded to travel from any such place within the state in which the trial is held, or

- (iii) requires disclosure of privileged or other protected matter and no exception or waiver applies, or
- (iv) subjects a person to undue burden.

## (B) If a subpoena

- (i) requires disclosure of a trade secret or other confidential research, development, or commercial information, or
- (ii) requires disclosure of an unretained expert's opinion or information not describing specific events or occurrences in dispute and resulting from the expert's study made not at the request of any party, or

(iii) requires a person who is not a party or an officer of a party to incur substantial expense to travel more than 100 miles to attend trial, the court may, to protect a person subject to or affected by the subpoena, quash or modify the subpoena or, if the party in whose behalf the subpoena is issued shows a substantial need for the testimony or material that cannot be otherwise met without undue hardship and assures that the person to whom the subpoena is addressed will be reasonably compensated, the court may order appearance or production only upon specified conditions.

## (d) DUTIES IN RESPONDING TO SUBPOENA.

(1) A person responding to a subpoena to produce documents shall produce them as they are kept in the usual course of business or shall organize and label them to correspond with the categories in the demand.

(2) When information subject to a subpoena is withheld on a claim that it is privileged or subject to protection as trial preparation materials, the claim shall be made expressly and shall be supported by a description of the nature of the documents, communications, or things not produced that is sufficient to enable the demanding party to contest the claim.

Schedule A

DOCUMENT REQUESTS

1. All Documents and things Concerning the prosecution, issuance, licensing, scope or validity of the '420 patent.
2. All Documents and things Concerning the prosecution, issuance, licensing, scope or validity of the '489 patent.
3. All Documents and things Concerning the interference declared at the USPTO between the U.S. Patent App. Ser. No. 09/506,160 and the '489 patent.
4. All Documents and things Concerning conception and reduction to practice of the invention(s) described and claimed in the '420 patent.
5. All Documents and things Concerning tests, data, and analyses underlying, or performed in connection with preparing Figures 1A, 1B, 3 and 4 and Table 1 in the '420 patent.
6. All Documents and things Concerning the '080 patent, including but not limited to its validity, or the scope of any of its claims.
7. All Documents and things Concerning tests and analyses on LYSO containing less than 15% Yttrium.
8. All Documents and things Concerning a comparison of the properties of LSO and LYSO.
9. All Communications with Saint-Gobain or Philips Concerning LSO or LYSO.
10. All Documents and things Concerning any drafts, revisions, or publications of any scientific articles, or scientific meeting presentations or posters, abstracts, or dissertations regarding LYSO and its properties.
11. All Documents and things Concerning the involvement of Siemens or Charles L. Melcher the development, manufacture, sale or offer for sale of any rare earth scintillation crystal, including but not limited to all Communications with Siemens or Charles L. Melcher Concerning the development, manufacture, sale or offer for sale of any rare earth scintillation crystal.
12. All Documents and things, including Communications with Saint-Gobain or Philips, Concerning this Litigation.
13. All Documents and things Concerning Saint-Gobain's License to the '420 patent, including Documents created in the course of preparing, negotiating, and entering into the License and Communications with Saint-Gobain.

### DEFINITIONS

As used in these requests, the following terms and phrases shall have the following definitions:

1. "Document" as used herein is defined to be synonymous in meaning and equal in scope to the usage of this term in Federal Rule of Civil Procedure 34(a), including, without limitation, electronic or computerized data compilations. A draft or non-identical copy is a separate Document within the meaning of this term.
2. "Concerning" as used herein means relating to, referring to, reflecting, describing, evidencing or constituting.
3. "Communication" as used herein means any transmittal of information (in the form of facts, ideas, inquiries, or otherwise).
4. "Siemens" as used herein means, individually and collectively, Siemens Medical Solutions, Inc., and any of its corporate parents, predecessors, successors, and past or present subsidiaries, affiliates, assigns, divisions, departments, officers, directors, principals, agents, representatives and employees. Siemens' predecessors include CTI, Inc. and CTI Molecular Imaging, Inc.
5. "Saint-Gobain" as used herein means, individually and collectively, Saint-Gobain Ceramics & Plastics, Inc., and any of its corporate parents, predecessors, successors, and past or present subsidiaries, affiliates, assigns, divisions, departments, officers, directors, principals, agents, representatives and employees.
6. "Philips" as used herein means, individually and collectively, Philips Medical Systems, and any of its corporate parents, predecessors, successors, and past or present subsidiaries, affiliates, assigns, divisions, departments, officers, directors, principals, agents, representatives and employees.
7. "LSO" as used herein means the single crystal form of cerium-doped lutetium oxyorthosilicate.
8. "LYSO" as used herein means the single crystal form of cerium-doped lutetium yttrium oxyorthosilicate.
9. "The '080 patent" as used herein means United States Letters Patent No. 4,958,080.
10. "The '420 patent" as used herein means United States Letters Patent No. 6,624,420.
11. "The '489 patent" as used herein means United States Letters Patent No. 6,323,489.
12. "Litigation" as used herein means Siemens Medical Solutions USA, Inc. v. Saint-Gobain Ceramics & Plastics, Inc., Case Number 07-190, before Judge Robinson in the District of Delaware.



13. "USPTO" as used herein means the United States Patent and Trademark Office.
14. "Saint-Gobain's License to the '420 patent" as used herein means the agreement titled "Patent License Agreement Between Research Foundation of the University of Central Florida and Saint-Gobain Ceramics & Plastics, Inc., Relating to Single Crystal Scintillators," which concerns the '420 patent.

IN THE UNITED STATES DISTRICT COURT  
FOR THE DISTRICT OF DELAWARE

SIEMENS MEDICAL SOLUTIONS USA,	)	
INC.,	)	
	)	
Plaintiff,	)	
	)	
v.	)	C.A. No. 07-190 (SLR)
	)	
SAINT-GOBAIN CERAMICS &	)	
PLASTICS, INC.,	)	
	)	
Defendant.	)	

**PLAINTIFF SIEMENS MEDICAL SOLUTIONS USA, INC.'S NOTICE  
OF DEPOSITION OF CRYSTAL PHOTONICS, INC. PURSUANT TO RULE 30(b)(6)**

In accordance with the provisions of Rules 26, 30 and 45 of the Federal Rules of Civil Procedure, Plaintiff Siemens Medical Solutions USA, Inc. ("Siemens"), hereby provides notice that commencing at 9:00 A.M. on March 5, 2008, at the offices of Hill Ward Henderson, 101 East Kennedy Boulevard, Suite 3700, Tampa, Florida 33602, or at such other time and place mutually agreed upon by counsel for the parties, it will take the deposition of Crystal Photonics, Inc. ("Crystal Photonics") by oral examination of witness(es) designated by Crystal Photonics to testify on its behalf as the person(s) most competent to testify concerning the matters listed in Attachment A. Pursuant to Federal Rule of Civil Procedure 30(b)(6), the witness(es) designated by Crystal Photonics should be prepared to testify as to such matters reasonably known or reasonably available to Crystal Photonics.

The depositions will be taken upon oral examination before an official authorized by law to administer oaths and will continue from day to day until completed. Pursuant to Rule 30(b)(2), testimony by the witness(es) will be recorded by stenographic and/or videographic means.

Dated: February 15, 2008

By: Charanjit Brahma

Gregg F. LoCascio  
Charanjit Brahma  
Sean M. McEldowney  
KIRKLAND & ELLIS LLP  
655 15<sup>th</sup> Street, N.W.  
Washington, DC 20005  
(202) 879-5000

Jack B. Blumenfeld (I.D. No. 1014)  
Maryellen Noreika (I.D. No. 3208)  
1201 North Market Street  
P.O. Box 1347  
Wilmington, DE 19899  
(302) 658-9200

*Attorneys for Plaintiff*  
**SIEMENS MEDICAL SOLUTIONS USA,  
INC.**

ATTACHMENT A

TOPIC 1: The '420 Patent, its prosecution, issuance, licensing, scope and validity, including without limitation the interference declared at the USPTO between the U.S. Patent App. Ser. No. 09/506,160 and the '489 patent.

TOPIC 2: Research regarding, and development and testing of, LYSO or LSO, including without limitation any testing of LYSO covered by any claim of the '420 Patent and any publications related thereto.

TOPIC 3: The properties and performance characteristics of LSO and LYSO.

TOPIC 4: All Documents Concerning any performance specifications for scintillation crystals received by Crystal Photonics or requests for proposal or requests for information about scintillation crystals to which Crystal Photonics has responded.

TOPIC 5: Any communications between Crystal Photonics and any other party regarding the Litigation or the '080 Patent.

TOPIC 6: All Documents and things related to the foregoing topics and Crystal Photonics' efforts to collect and produce Documents and things in response to the subpoena served by Siemens upon Crystal Photonics.

TOPIC 7: All Persons known to have knowledge of the foregoing topics other than knowledge derived solely from preparing a response to the subpoena served by Siemens upon Crystal Photonics.

DEFINITIONS

As used in the aforementioned topics, the following terms and phrases shall have the following definitions:

1. "Document" as used herein is defined to be synonymous in meaning and equal in scope to the usage of this term in Federal Rule of Civil Procedure 34(a), including, without limitation, electronic or computerized data compilations. A draft or non-identical copy is a separate Document within the meaning of this term.
2. "Concerning" as used herein means relating to, referring to, reflecting, describing, evidencing or constituting.
3. "Communication" as used herein means any transmittal of information (in the form of facts, ideas, inquiries, or otherwise).
4. "Crystal Photonics" as used herein means, individually and collectively, Crystal Photonics, Inc., and any of its corporate parents, predecessors, successors, and past or present subsidiaries, affiliates, assigns, divisions, departments, officers, directors, principals, agents, representatives and employees.
5. "LSO" as used herein means the single crystal form of cerium-doped lutetium oxyorthosilicate.
6. "LYSO" as used herein means the single crystal form of cerium-doped lutetium yttrium oxyorthosilicate.
7. "The '080 patent" as used herein means United States Letters Patent No. 4,958,080.
8. "The '420 patent" as used herein means United States Letters Patent No. 6,624,420.
9. "The '489 patent" as used herein means United States Letters Patent No. 6,323,489.

10. "Litigation" as used herein means Siemens Medical Solutions USA, Inc. v. Saint-Gobain Ceramics & Plastics, Inc., Case Number 07-190, before Judge Robinson in the District of Delaware.

11. "USPTO" as used herein means the United States Patent and Trademark Office.

# EXHIBIT 6

**FOLEY**  
FOLEY & LARDNER LLP

February 29, 2008

VIA E-MAIL, FACSIMILE AND U.S. MAIL

ATTORNEYS AT LAW  
WASHINGTON HARBOUR  
3000 K STREET, N.W., SUITE 500  
WASHINGTON, D.C. 20007-5143  
202.672.5300 TEL  
202.672.5399 FAX  
foley.com

WRITER'S DIRECT LINE  
202.672.5332  
kkrosin@foley.com EMAIL

CLIENT/MATTER NUMBER  
999200.0692

Charanjit Brahma, Esquire  
Kirkland & Ellis LLP  
655 Fifteenth Street, N.W.  
Washington, D.C. 20005

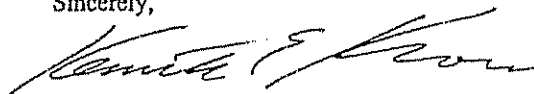
Re: *Siemens Medical Solutions USA, Inc. v. Saint-Gobain Ceramics & Plastics, Inc.*, (D. Del. C.A. No. 07-190-SLR)

Dear Mr. Brahma:

Enclosed herewith are the "Responses and Objections of Non-Party Leon Radomsky to Subpoena Served By Plaintiff." Pursuant to paragraph 5 therein, Mr. Radomsky has waived the attorney-client privilege and work-product immunity with respect to "the non-infringement opinion ("Opinion"), dated March 16, 2006 rendered by Foley on behalf of Saint-Gobain Ceramics & Plastics Inc. (Saint-Gobain"), and Foley's files created and maintained specifically in connection with the production of the Opinion."

Copies of documents based upon this waiver were provided to you, Bates Nos. SGCP092122-093201, by Mr. John C. Ohman, Saint-Gobain's counsel, by letter dated January 23, 2008. To avoid duplication, Mr. Radomsky will be relying on that production in response to the corresponding document requests in your subpoena.

Sincerely,



Kenneth E. Krosin

KEK:krb  
Enclosure

cc: John C. Ohman, Esquire

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# EXHIBIT 7

**REED FAX**  
FOR PATENT CONNECTION

# United States Patent File History

## Tab Listings

- A. Differences (if applicable)
  - A1. US References
  - A2. Foreign References
- B. Jacket (back of file, contents flap, index of claims, PTO 270, searched)
- C. Printed Patent
- D. Specification (serial no, sheet abstract, specification, claims)
- E. Oath
  - E1. Small entity status (if applicable)
- F. Drawing Figures (if applicable)
- G. PTO/Applicant Correspondence
- H. Original Patent Application (in cases of FWC)

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SMS003145

SERIAL NUMBER <b>07/389502</b>		PATENT DATE <b>SEP 18 1990</b>		PATENT NUMBER <b>4958080</b>	
SERIAL NUMBER <b>07/389,552</b>	FILING DATE/CLASS <b>10/04/89</b>	SUBCLASS <b>250</b>	GROUP/ART. UNIT <b>301.4</b>	EXAMINER <b>258</b>	EXAMINER <b>111</b>
APPLICANT <b>CHARLES I. PELCHER, WEST HEDDING, CT.</b>					
<p>***CONTINUING DATA*****</p> <p>VERIFIED THIS APPLN IS A CON OF <b>07/254,353 10/05/88</b></p>					
<p>***FOREIGN/PCT APPLICATIONS*****</p> <p>VERIFIER</p>					
<p>Foreign priority claimed <input type="checkbox"/> Yes <input checked="" type="checkbox"/> No</p> <p>35 USC 119 conditions met <input type="checkbox"/> Yes <input checked="" type="checkbox"/> No</p> <p>AS FILED <input checked="" type="checkbox"/> CT</p> <p>STATE OR COUNTRY <b>CT</b></p> <p>SHEET DRAWS <b>2</b></p> <p>TOTAL CLAIMS <b>12</b></p> <p>INDEX CLAIMS <b>4</b></p> <p>FILING FEE RECEIVED <b>526.00</b></p> <p>ATTORNEY'S DOCKET NO. <b>275151539151</b></p>					
<p>Verified and Acknowledged <b>EXAMINER'S OFFICE</b></p> <p><b>DRUMBAUGH, GRAVES, DONOHUE &amp; RAYMOND</b></p> <p><b>30 ROCKEFELLER PLAZA</b></p> <p><b>NEW YORK, NY 10112</b></p>					
<p><b>LUTETIUM COMPOSITE SINGLE CRYSTAL SCINTILLATOR DETECTOR</b></p> <p><b>CERTIFICATE</b></p> <p><b>JUN 9 1992</b></p> <p><b>OF CORRECTION</b></p> <p><i>1 sheet of drawing transferred from S.N. 254,353 2/26/89</i></p>					
<p>PARTS OF APPLICATION FILED SEPARATELY</p>					
NOTICE OF ALLOWANCE MAILED <b>MAR -1 1990</b> <b>5-8-90</b>		PREPARED FOR ISSUE <b>8/1/90</b>		CLAIMS ALLOWED	
ISSUE FEE Amount Due <b>\$620.00</b> Date Paid <b>7/16/90</b>		Assistant Examiner <b>D. Fitch</b>		Total Claims <b>2</b>	Print Claim <b>1</b>
<p><b>JACK COOPER</b></p> <p><b>PRIMARY PATENT EXAMINER</b></p> <p><b>ART UNIT 113</b></p> <p><b>Primary Examiner</b></p>		<p><b>DRAWING</b></p> <p>Sheets Drawn <b>2</b></p> <p>Figs Drawn <b>2</b></p> <p>Print Figs <b>NONE</b></p>			
<p><b>ISSUE CLASSIFICATION</b></p> <p>Class <b>250</b></p> <p>Subclass <b>483.1</b></p>		<p><b>ISSUE BATCH NUMBER</b></p> <p><b>F33</b></p>			
<p><b>WARNING:</b> The information disclosed herein may be restricted. Unauthorized disclosure may be prohibited by the United States Code Title 35, Sections 122, 181 and 306. Possession outside the U.S. Patent &amp; Trademark Office is restricted to authorized employees and contractors only.</p>					

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## CONTENTS

Received  
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1.	Application _____ papers.	
2.	45 Day Letter (152)	AUG 18 1989
3.	Statement (152)	9-11-89 OK
4.	<i>Per A. L. A.</i>	<i>Feb. 20, 1990</i>
5.	<i>Disclosure Statement</i>	<i>Nov. 6, 1989</i>
6.	<i>Ex. D. Interview</i>	<i>Feb. 2, 1990</i>
7.	<i>Per A. L. B.</i>	MAR - 1 1990
8.	ACCERS ACKNOWLEDGEMENT	MAY 02 1990
9.	<i>Notice of Allegations</i>	5-8-90
10.	Statement to DOE	MAY 16 1990
11.	DOE Action & Waiver	JUL 22 1990
12.	PTO GRANT SEP 16 1990	
13.	<i>Disclosure Statement</i>	<i>12-5-89</i>
14.	<i>Reg for Conf. Rule 322</i>	<i>10/9/90</i>
15.	<i>Director's Report</i>	<i>5/4/92</i>
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## INDEX OF CLAIMS

Claim		Date	
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Class	Sub.	Date	Exmr.
252	301.4F	2/24/00	JF
250	483.1		
	361R		

SEARCH NOTES		
	Date	Exmr.

INTERFERENCE SEARCHED			
Class	Sub.	Date	Exmr.
252	301.4	2/24/00	JF
250	361R		
	483.1		

SMS003149

PATENT NUMBER		ORIGINAL CLASSIFICATION		AU236 CAF	
APPLICANT'S SERIAL NUMBER		CLASS	SUBCLASS		
389502		250	41831		
APPLICANT'S NAME (PLEASE PRINT)		CROSS REFERENCE(S)			
Melcher		CLASS	SUBCLASS	FOR SUBCLASS PERIOD	
		250	361R		
		252	3014-F		
IF FOREIGN, ORIGINAL PATENT NUMBER					
INTERNATIONAL CLASSIFICATION (INT. CL. 4)					
DDAIX	11/79				
GDIJ	1/58				
		GROUP	ASSISTANT EXAMINER (PLEASE STAMP OR PRINT FULL NAME)		
		113	JACK COOPER		
ISSUE CLASSIFICATION SLIP		U.S. DEPARTMENT OF COMMERCE PATENT AND TRADEMARK OFFICE			

SMS003150



## United States Patent [39]

Melcher

[31] Patent Number: 4,958,080

[45] Date of Patent: Sep. 18, 1990

[54] LUTETIUM ORTHOSILICATE SINGLE CRYSTAL SCINTILLATOR DETECTOR

[75] Inventor: Charles L. Melcher, West Redding, Conn.

[73] Assignee: Schlumberger Technology Corporation, New York, N.Y.

[21] Appl. No.: 389,507

[22] Filed: Aug. 4, 1989

Related U.S. Application Data

[63] Continuation of Ser. No. 754,111, Oct. 6, 1978, abandoned.

[31] Int. Cl.<sup>3</sup> ..... C09K 11/79; G01J 1/38

[32] U.S. Cl. .... 250/483.1; 250/361.R;

252/381.4-P

[38] Field of Search ..... 252/381.4 P; 250/483.1;

250/361 R

[36] References Cited

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4,201,611 6/1980 Yamabe et al. .... 252/381.4 P

4,641,711 9/1981 Takagi et al. .... 252/381.4 P

## FOREIGN PATENT DOCUMENTS

51-57019 5/1976 Japan ..... 252/381.4 P

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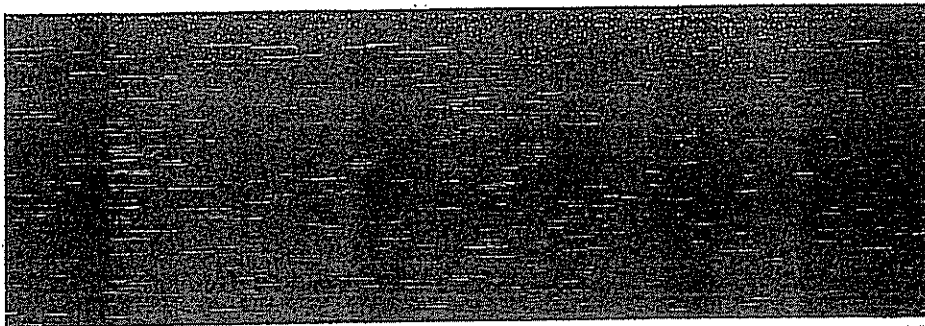
"Serie Isomorphes d'Orthosilicates ( $\text{Y}_2\text{SiO}_5$ ) et d'Orthosilicates ( $\text{Y}_2\text{GeO}_5$ ) De Terres Rares", Dubessy et al., *Mat. Res. Bull.*, vol. 3, pp. 193-198, 1968."Preparation and Cathodoluminescence of  $\text{Ce}^{3+}$ -Activated Yttrium Silicates and Some Isomorphous Compounds", A. H. Gomes de Mesquita et al., *Mat. Res. Bull.*, vol. 4, pp. 643-650, 1969."Growth of Lanthanide Orthosilicate Single Crystals and Their Structural and Optical Characteristics", G. V. Anisova et al., Translated from *Izvestiya Akade-*mii Nauk SSSR, *Neorganicheskoe Materialy*, vol. 17, No. 6, pp. 1037-1042, Jan. 1981."Site Selectively Doped Luminescence of  $\text{Eu}^{3+}$  in Gadolinium, Yttrium and Lanthanum Orthosilicates", Hoira et al., *Journal of the Less-Common Metals*, 125 (1986) 215-220."Cerium-Activated  $\text{Gd}_2\text{SiO}_5$  Single Crystal Scintillator", Takagi et al., *Appl. Phys. Lett.*, vol. 42, No. 1, 1 Jan. 1983, pp. 43-45."Controlled Growth of Rare-Earth Orthosilicates ( $\text{Ln}_2\text{SiO}_5$ )", Brundillo et al., *Journal of Crystal Growth* 79 (1986) pp. 306-315."Sol-Gel Deposition of  $\text{Tb}^{3+}/\text{Y}_2\text{SiO}_5$  Cathodoluminescent Layers", Balkevich et al., *Am. Ceram. Soc. Bull.* 66(10) 1505-09 (1987)."Characteristics Luminescence", Blanc et al., *Philips Technical Review*, vol. 31, 1970, No. 10."Development of New Scintillation Crystal  $\text{Gd}_2\text{SiO}_5$  Crystals for Gamma-Ray Detection", Ishii et al., submitted to 4th Experts Meeting on Position CT between AIST Japan and STU Sweden, Stockholm, Mar. 17-20, 1986.Primary Examiner—Jack Cooper  
Attorney, Agent or Firm—Brimbaugh Graves Donohue & Raymond

[57]

## ABSTRACT

A scintillator for use as a gamma ray or beta radiation detector is composed of a single crystal of cerium-activated lutetium orthosilicate having the general formulation  $\text{Ce}_x\text{Lu}_{1-x}\text{SiO}_5$ . In a borehole logging application, the detector is mounted in a logging sonde with a high energy neutron source, for movement through a borehole traversing earth formations. Gamma radiation from the surrounding formations is detected and analyzed to provide information concerning hydrocarbons in the formations.

1 Claim, 1 Drawing Sheets



SMS003151



U.S. Patent Sep. 18, 1990 Sheet 1 of 2 4,958,080

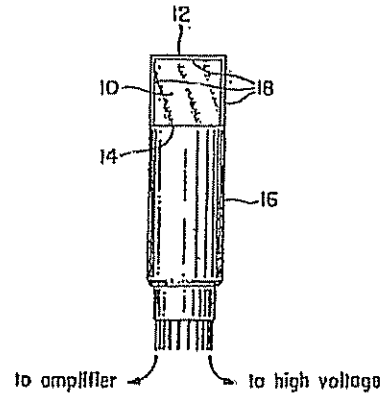
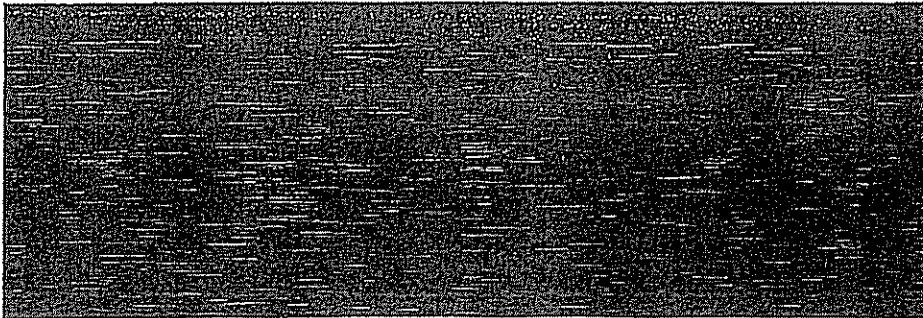
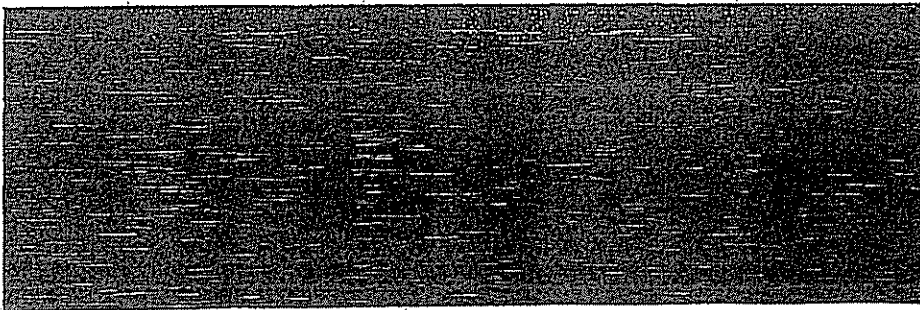
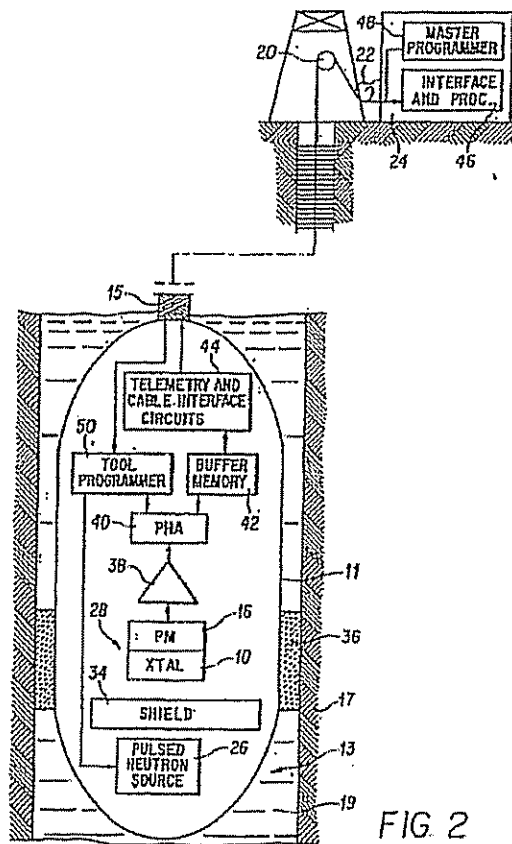


FIG. 1



SMS003152

U.S. Patent Sep. 18, 1990 Sheet 2 of 2 4,958,080



SMS003153

4,958,080

# LUTETIUM ORTHOSILICATE SINGLE CRYSTAL SCINTILLATOR DETECTOR

## BACKGROUND OF THE INVENTION

This application is a continuation, now abandoned of application Ser. No. 254,353, filed Oct. 6, 1988.

The present invention relates to a single crystal scintillation detector for gamma rays and like radiation and, more particularly, to a single crystal scintillation detector composed of lutetium orthosilicate.

A well-known form of detector for gamma rays and like radiation (such as x-rays, cosmic rays, and energetic particles of approximately 1 MeV and above) employs a transparent single crystal, known as a scintillator, which responds to impinging radiation to emit light pulses. The light pulses are optically coupled to the input of a photomultiplier tube, which generates a voltage signal related to the number and amplitude of the light pulses received. Scintillators of this class have found wide application in various fields, such as nuclear medicine, physics, chemistry, mineral and petroleum exploration, etc.

Perhaps the most widely used type of scintillator is thallium-doped sodium iodide, NaI (Tl). Relatively inexpensive to produce and capable of providing a high light output in response to impinging radiation, NaI detectors have found general use, for example, in logging tools for oil well logging operations, where either naturally occurring or induced gamma radiation is detected to aid in the location of petroleum deposits.

Other known single crystal scintillators used for gamma ray detection include cesium iodide (cesium or thallium activated) and bismuth germanate (BGO). Organic scintillators, such as anthracene, anthracene, stilbene and similar materials, have also been employed, particularly where very high count rates are important, although they generally are not as useful as inorganic scintillators for the detection of gamma rays.

All of the foregoing types of scintillators have one or more disadvantages as gamma ray detectors. For example, NaI scintillators have comparatively low density, and therefore low radiation detection efficiency, slow scintillation decay and a large and persistent afterglow, which impairs counting rate performance and lead to pulse pile-up, and are hygroscopic. Although BGO scintillators do not suffer from the low density and hygroscopicity problems of NaI scintillators, they do have a relatively slow scintillation decay time and low light output, which drops still lower at higher temperatures. The index of refraction of BGO is also relatively high, resulting in light loss by internal reflections. These and other disadvantages of known scintillators have limited their usefulness as gamma ray detectors or have imposed restrictions on the manner of their use. In the latter case, such as in logging, for example, restrictions in logging speed, statistical reliability, tool size and the like have resulted directly from the scintillator material employed in a given logging tool. Accordingly, efforts have long continued to develop still more useful and reliable scintillation detectors and gamma ray scintillation detectors in particular.

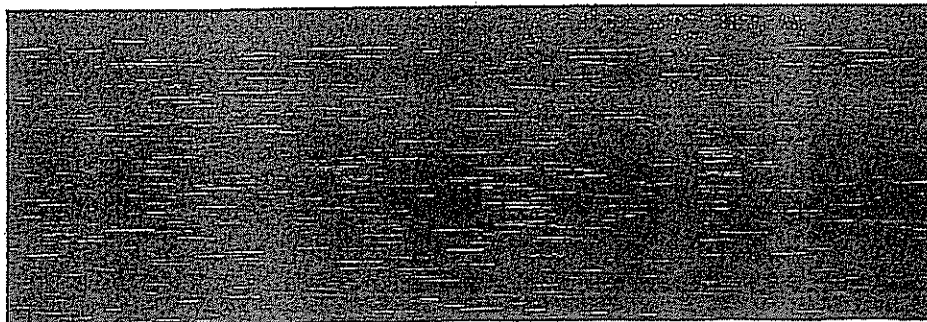
More recently, a gamma ray detector employing a scintillator formed of a single crystal of cerium-activated gadolinium orthosilicate (GSO) has been proposed. The GSO scintillator has the advantages as a gamma ray detector of high effective atomic number,

high density, fast scintillation decay, relatively low index of refraction, but has the disadvantages of low light output, a strong tendency to cleave which makes cutting and polishing difficult, and, more significantly, very high thermal neutron capture cross section (49,000 barns). This last characteristic strongly suggested that GSO scintillators would have very limited utility, if any, in those applications, such as many nuclear well logging tools for instance, where the gamma radiation to be detected is induced by neutron irradiation. This is because gadolinium, upon the capture of thermal neutrons, emits gamma radiation which would interfere with the detection of the external gamma rays of interest.

Such a GSO scintillator detector is described in U.S. Pat. No. 4,647,781, issued Mar. 3, 1987, for use in positron computed tomography. There is no disclosure in the '781 patent, however, of the suitability of the GSO scintillator as a gamma ray detector in the harsh conditions of borehole logging. Indeed, it was the present inventor and his co-workers who, through experimentation and evaluation, first discerned that the GSO scintillator was useful as a borehole detector. This invention is disclosed in the copending, commonly-owned U.S. application Ser. No. 149,953 filed Feb. 2, 1988 by Charles L. Melcher et al., which is a continuation of U.S. application Ser. No. 812,229 filed Dec. 23, 1985. U.S. application Ser. No. 149,953 issued as U.S. Pat. No. 4,883,956, granted Nov. 28, 1989.

Success in adapting the GSO scintillator to borehole use led the inventor to consider other rare earth compounds as possible scintillators for gamma ray (and the like) detection. As a first step, various phosphor materials were studied in powder form for initial evaluation in order to identify potential candidates for crystal growth. This initial evaluation was done using procedures similar to those described by A. E. Goeta de Mesquita et al. at pages 649-650 of Materials Research Bulletin, Vol. 4, No. 9, 1969, Pergamon Press, Inc., who were investigating the properties of rare-earth phosphors as their primary goal. The term phosphor as used in the Goeta de Mesquita et al. paper and as used herein refers to the fine powdered form. Such phosphors are typically used as coatings on cathode ray tube screens, fluorescent light bulbs, and the like, where they convert impinging electrons or ultraviolet radiation into visible light pulses. They are, however, not suitable as gamma ray or like radiation detectors since energetic photons or particles have high probability of passing through the thin coating with no interaction. If the coating is made sufficiently thick to stop gamma rays, the resulting opacity of the phosphor layer would trap most of the scintillation signal. Consequently, only transparent single crystals are useful as gamma ray detectors.

Although none of the physical and optical properties of the rare earth phosphors were known, their scintillation properties were not known and could not be predicted in advance because of the complex and only partially understood nature of the scintillation mechanism. For example, on the basis of atomic number, ionic radius, electronic charge, density, refractive index, and absence of absorption bands, cerium-doped ytterbium orthosilicate powder and cerium-doped lutetium orthosilicate powder would be expected to have very similar scintillation properties. In fact, however, the phosphor experiments showed that ytterbium orthosilicate has 1000 times less scintillation output compared to lutetium orthosilicate.



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Moreover, it was not at all clear whether the luminescence properties or other properties of the single crystal form of a rare earth compound would be the same as those of the phosphor form. Generally, in fact, the scintillation properties of single crystal scintillators are not readily determinable or predictable with certainty in advance from the phosphor.

#### SUMMARY

In accordance with the present invention, therefore, an improved scintillator for use as a gamma ray (or the radiobeam) detector is provided which comprises a single crystal of cerium-activated lutetium orthosilicate having the general formulation  $\text{Ce}_x\text{Lu}_{1-x}\text{SiO}_4$ . Generally, the value of  $x$  (as measured in the initial melt from which the crystal is pulled) may be varied within the approximate range of from 0.001 to 0.1, with the preferred range of  $x$  being from approximately 0.005 to 0.015. When assembled in a complete detector, the scintillator crystal is optically coupled, either directly or through a suitable light path, to the photosensitive surface of a photodetector for generation of an electrical signal in response to the excitation of a light pulse by the scintillator. The LSO scintillator of the invention possesses certain important characteristics, most notably high light output, very short decay time and high detection efficiency, that make it superior to prior scintillators as a gamma ray or like radiation detector, particularly in the borehole logging environment.

#### BRIEF DESCRIPTION OF THE DRAWING

FIG. 1 is a diagrammatic view of one embodiment of an LSO scintillation detector in accordance with the present invention; and

FIG. 2 is a schematic diagram of a borehole logging probe in which the LSO scintillation detector of FIG. 1 may be used.

#### DESCRIPTION OF REPRESENTATIVE EMBODIMENT

For illustrative purposes, a representative embodiment of the invention is described hereinafter in the context of a gamma ray detector. It will be understood, of course, that the utility of the novel LSO single crystal scintillator of the invention is not limited to the detection of gamma radiation but that it has general application to the detection of other types of like radiation, e.g., X-rays, cosmic rays, and energetic particles.

In FIG. 1, a single crystal LSO scintillator 10 is shown encased within the housing 12 of a gamma ray detector. One face 14 of the scintillator is placed in optical contact with the photosensitive surface of a photomultiplier tube 16. Alternatively, the light pulses could be coupled to the photomultiplier via light guides or fibers, lenses, mirrors, or the like. The photomultiplier can be replaced by any suitable photodetector such as a photodiode, microchannel plate, etc. In order to direct as much of each light flash to the photomultiplier as possible, the other faces 18 of the scintillator are preferably wrapped or covered with a reflective material, e.g., Teflon tape, magnesium oxide powder, aluminum foil, or titanium chloride paint. Light pulses emitted by the LSO crystal upon the incidence of radiation are intercepted, either directly or upon reflection from the surfaces 18, by the photomultiplier, which generates electrical pulses or signals in response to the light pulses. These electrical output pulses are typically first amplified and then subsequently processed as desired, e.g., in a pulse height amplifier, to obtain the parameter of interest regarding the detected radiation. The photomultiplier is also connected to a high voltage power supply, as indicated in FIG. 1. Other than the LSO scintillator, all of the components and materials referred to in connection with FIG. 1 are conventional, and thus need not be described in detail.

As the first step in the evaluation of cerium-activated lutetium orthosilicate  $\text{Ce}_x\text{Lu}_{1-x}\text{SiO}_4$  as a scintillator, the material was synthesized in phosphor (powder) form and the scintillation properties of the resulting compound were analyzed. From this analysis, it was determined that LSO was sufficiently promising as a scintillator material to warrant the additional effort and expense of growing in single crystal form. LSO was chosen as a candidate for phosphor synthesis because of its high atomic number and high density and the absence of optical absorption bands in the visible part of the spectrum. The principal physical and optical properties of LSO are compared to those of GSO and NaI (Tl) in Table 1, from which it may be seen that LSO appears to offer advantages over GSO in all properties and over NaI (Tl) in the areas of atomic number, density, index of refraction and hygroscopicity.

TABLE 1

	NaI(Tl)	GSO	LSO
Diffractive streak number	31	29	44
Density (gm/cm <sup>3</sup> )	3.67	4.71	7.4
Index of Refraction	1.83	1.71	1.82
Refractive index surface (nm)	4.3	4,000	44
Hygroscopicity	Yes	No	No

As Ce-activated single crystals of LSO had not previously been grown, the scintillation properties of single-crystal LSO were unknown. Also unknown, therefore, was whether the properties of the LSO phosphor would be retained when the same material was grown into a single crystal.

The phosphor synthesis procedure consisted of the following steps:

(1) Fine powders of  $\text{Lu}_2\text{O}_3$  ( $\text{Lu}=\text{Ce}, \text{La}$ )  $\text{SiO}_2$  and either  $\text{CeO}_2$  or  $\text{Ce}_2\text{O}_3$  in the appropriate atomic ratios were mechanically milled and loaded into an alumina boat. In each case the molar concentration of Ce was 0.5% relative to Lu or La.

(2) Each of the four compositions was sintered at 1500° C. for four hours in a tube oven in one of three atmospheres: neutral (pure argon), reducing (argon +2% hydrogen), or oxidizing (air). When a reducing atmosphere was used, a high purity alumina furnace tube was employed because the standard mullite tube was found to react with hydrogen at high temperature and contaminate the samples. The resulting material was solid and was ground into a powder for analysis.

(3) The fluorescence emission of the powder was excited with ultraviolet light and the emission and excitation spectra were recorded with a Spex Fluorolug (Model 212) spectrophotometer. For emission measurements above room temperature, the sample chamber was continuously flushed with argon to suppress spurious fluorescence emission which occurred when the sample was heated above 100° C. to air.

Table 2 shows the intensity of the fluorescence emission measured at room temperature.

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TABLE 2

	GdO:Co <sup>2+</sup>	GdO:Co <sup>2+</sup> :Ce <sup>3+</sup>	LSO:Co <sup>2+</sup>	LSO:Co <sup>2+</sup> :Ce <sup>3+</sup>
Atmos + H <sub>2</sub>	44	77	71	77
Air	1.8	1.6	11	23
Argon	12	2.5	122	14

(4) The fluorescence decay times of the phosphors were measured by spreading a thin layer of the phosphor on the face of a fast photomultiplier (Amperex 2000Q) and exciting the emission with an <sup>241</sup>Am gamma-ray source. The pulse shape was observed at the anode of the photomultiplier with a fast oscilloscope and the decay time estimated from the oscilloscope trace.

Although the synthesis conditions employed were not necessarily optimum for each phosphor, inasmuch as the emission intensity of LSO was greater than that of GSO under all six synthesis conditions (three different atmospheres and two forms of cerium), it may be concluded that LSO's scintillation efficiency is intrinsically greater. The scintillation mechanism in cerium-activated phosphors is generally assumed to involve electronic transitions (5d to 4f) within the Ce<sup>3+</sup> ion. For this reason, it was expected that the most efficient phosphors would result from using Co<sup>2+</sup> (i.e., Co<sup>2+</sup>) as a starting material and performing the synthesis in a slightly reducing atmosphere. (Ar+H<sub>2</sub>) is order to maintain the cerium in the +3 charge state. Surprisingly, as Table 2 shows, in every case except LSO prepared in air the best phosphors were those in which Co was added as CoO<sub>2</sub>. Also note that although the best GSO phosphors were those prepared in a reducing atmosphere, the best LSO phosphors were those prepared in a neutral atmosphere. Synthesis in air resulted in phosphors with lower light output and, more importantly, with emission spectra that differed from the single crystal emission spectrum.

Table 3 summarizes the scintillation properties of GSO and LSO phosphors, synthesized in the foregoing manner:

TABLE 3

	GdO:Co <sup>2+</sup>	LSO:Co <sup>2+</sup>
Light output	7	15-10
Decay time	50 ns	50 ns
Emission peak	430 nm	413 nm
Temperature response	-24%/°C	-13%/°C

A comparison of the physical properties (Table 7) and the scintillation properties (Table 3) of GSO and LSO show LSO to be superior to GSO in most areas. The light output of LSO is a factor of 1.5 to 10 greater than GSO depending on the synthesis conditions. This is a particularly notable advantage over GSO. A comparison of the emission spectra of the GSO:Co<sup>2+</sup> and LSO:Co<sup>2+</sup> phosphors synthesized in argon (with the emission light wavelength corresponding to the strongest emission band for each phosphor, 343 nm for GSO and 355 nm for LSO) shows the LSO emission to

have a different shape and to be shifted somewhat towards the shorter wavelengths than the GSO emission. This apparent shift of the LSO spectrum, if exhibited in the single crystal form, would be advantageous since it would result in an emission that better matches the spectral response of high temperature photomultipliers.

The scintillation decay time of LSO of about 50 ns compares favorably with GSO's 60 ns. The effective atomic number of LSO is 66 compared to 59 for GSO and the density of LSO is 7.4 gm/cc compared to 6.7 gm/cc for GSO, both of which contribute to a higher radiation detection efficiency for LSO. The index of refraction of LSO is 1.81 compared to 1.91 for GSO, which results in less trapping of scintillation light. LSO is very much less sensitive to neutrons, since the thermal cross section for La is 77 barns compared to 47,000 barns for Gd.

The temperature response of LSO is somewhat worse than GSO. At 150° C, GSO's light output decreases to about 65% of its room temperature value, while LSO's light output decreases to about 70% of its room temperature value at 150° C. On the other hand, the emission spectrum of LSO was found to shift somewhat to shorter wavelengths than the GSO emission as temperature increased from room temperature up to 175° C (the highest temperature measured). Again this would be advantageous in the single crystal form in terms of matching photomultiplier response.

Lanthanum has a radioactive isotope (<sup>138</sup>La) that produces a background noise level in the crystal. This could be eliminated by growing the crystal from pure <sup>139</sup>La, or it could be handled by conventional background subtraction techniques.

The emission spectrum of LSO exhibits three bands (452 nm, 298 nm, and 313 nm) and is similar to the GSO emission spectrum except that the bands are shifted to somewhat longer wavelengths.

Although the comparison of the LSO and GSO phosphors showed LSO to be a promising scintillator for use in a gamma ray detector, the properties of the single crystal form cannot be predicted with certainty. Light output, one of the most important properties of a single crystal scintillator, is especially difficult to predict from the phosphor form. In order to continue with a more detailed evaluation of the material as well as to test a practical detector, therefore, it was necessary to grow a single crystal form of LSO. This was done from melts by the conventional Czochralski method, as described, for example, by C. D. Ryan et al., "Czochralski Growth of Rare-Earth Orthosilicates (Ln<sub>2</sub>SiO<sub>5</sub>)", *Journal of Crystal Growth*, No. 79, pp. 304-315, 1986.

Table 4 summarizes the scintillation properties of the LSO single crystals grown, as selected and cut to minimize imperfections. The composition of the melts was Ce<sub>x</sub>(La<sub>1-x</sub>)<sub>2</sub>SiO<sub>5</sub>, where x is the decimal value of the percentage set out in Table 4 under the heading "Ce concn". Cerium concentration in the crystals was on the order of 20%-30% of that in the melt.

TABLE 4

Size (mm)	Crystal defects	Ce concn (wt. %)	Light output (mW/cm <sup>2</sup> )	Energy res. %	Decay constant (ns)	Emission peak (nm)	Crystals max (gamma ray) resolution (nm)
1 3 X 4 X 1	none	1.0	154	2.8	44	373	413
2 7 X 9 X 1	none	1.3	109	12	61	373	413
3 5 X 3 X 25	none	0.7	114	11.3	61	374	413

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TABLE 4-continued

the (mm)	color	defects	Ca conc. (%)	light output (cd/m <sup>2</sup> )	energy res. (keV)	decay constant (s <sup>-1</sup> )	relative res. (TV monitor) (%)	relative res. (gamma detector) (%)
8	3 x 3 x 1	none	few	0.8	101	17	14	313

Crystals 1, 2 and 4 were cut to the sizes listed from larger single crystals (5 mm x 6 mm x 28 mm, 7 mm x 9 mm x 27 mm and 8 mm x 8 mm x 33 mm, respectively), but crystal 3 was the original size. All were clear of

within the approximate range of from 0.1% to 10%, i.e., 0.0015 x 0.1. The preferred melt cation concentration is within the range of from approximately 0.3% to 1.5%, i.e., 0.0035 x 0.015.

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from capture within the crystal is greatly reduced relative to GSO. Finally, the radiation length of LSO is as good as that of BGO and considerably shorter than either GSO or NaI, with consequent advantages in the crystal size required.

As may be appreciated from Tables 4 and 5, the LSO single crystal scintillator is comparable to or exceeds other known scintillators in nearly all properties important for use as a gamma ray detector, i.e., light output, energy resolution, efficiency of detection of high energy photons, scintillation decay time, hygroscopicity, susceptibility of crystal to mechanical damage, reflective tube response, transparency of the crystal to its own scintillation emission, and absence of induced gamma radiation within the crystal. The only area in which LSO compares unfavorably is to the temperature sensitivity of the gamma-ray excited emission. In controlled environments, e.g., laboratories, hospitals, etc., this presents no problem. Even in high temperature environments, moreover, the unique combination of light output, speed, and detection efficiency found in LSO will in many instances justify the additional cost of isolating the scintillator from environmental temperature changes.

As discussed above, the LSO scintillator detector of the present invention is particularly effective as a radiation detector in a borehole logging environment, such as for petroleum exploration. In such use, the detector forms part of a logging system which may be of the type disclosed in the aforementioned copending U.S. applications Ser. No. 149,953 and illustrated in FIG. 1 herein.

FIG. 1 shows a logging sonde 11 for sensing gamma radiation resulting from bombardment of a formation with high energy neutrons and detecting the energy of the radiation for subsequent spectral analysis. The sonde 11 is supported in a borehole 13 on an armored multiconductor cable 15. The borehole 13 traverses a formation 17 and is filled with fluid 19, and may be open as shown or cased. The sonde 11 as described below may be constructed in accordance with U.S. Pat. No. 4,317,993 to Hertog, Jr. et al., assigned to the assignee of the present application. The sonde 11 is moved in the borehole 13 by playing the cable 15 out and reeling it back in over a sheave wheel 20 and a depth gauge 22 by means of a winch forming part of a surface equipment 24. Usually the logging measurements are actually made while the sonde 11 is being raised back up the borehole 13, although in certain circumstances they may be made on the way down instead or as well.

The sonde 11 includes a pulsed neutron source 26 for producing primary radiation to bombard the formation 17 with fast neutrons as the sonde 11 travels up the borehole 13, and a radiation detector 28 for detecting secondary (gamma) radiation induced thereby in the borehole 13 and the formation 17. The neutron source 26 is preferably of the pulsed accelerator type described in U.S. Pat. Nos. 3,461,701 to Goodson and 3,546,512 to Finnerty, both commonly owned with this application. This type of source is particularly suited to the generation of discrete bursts of high energy or fast neutrons, e.g., at 14 MeV, with a controlled duration and repetition rate.

The detector 28 is of a type appropriate to the detection of gamma radiation and the production of an electrical signal corresponding to each detected gamma ray and having an amplitude representative of the energy of the gamma ray. To this end the detector 28 is as shown

in FIG. 1, including a cerium-activated LSO scintillation crystal 30 optically coupled to a photomultiplier tube (PMT) 32. Suitable tubes are manufactured by EMR Photocentric, Princeton, N.J.

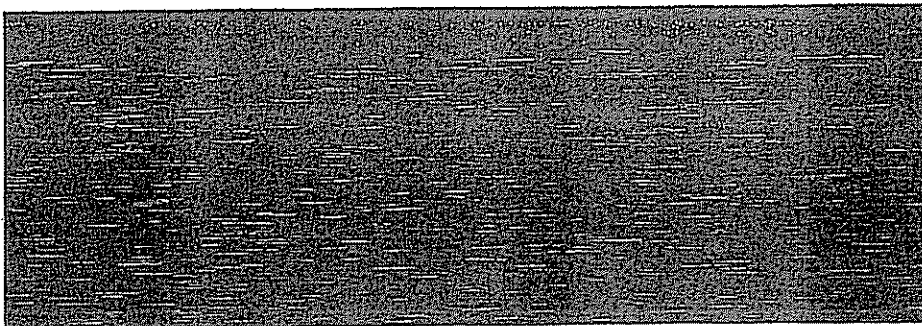
Although not as important as in the case of a GSO scintillator, a neutron shield 34 may be located between the source 26 and the detector 28 to limit direct bombardment of the detector 28 by neutrons from the source 26, thereby avoiding saturation of the detector 28 by such direct irradiation. In addition, especially in the case of measurement of capture gamma radiation, the sonde 11 may be surrounded by a sleeve 36 impregnated with boron carbide and located in the general vicinity of the source 26 and the detector 28. This sleeve displaces borehole fluid in the region of the detector 28, and absorbs neutrons scattered by the formation towards the detector 28, without significantly attenuating gamma radiation emanating from the formation. The net effect is to reduce the possibility of neutron interactions with the borehole contents and the material of the sonde 11 in proximity to the detector 28 and which would otherwise produce detectable gamma rays constituting an undesirable perturbation of the required gamma ray measurement.

Electrical power for the sonde 11 is supplied via the cable 15 from the surface equipment 24. The sonde 11 includes power conditioning circuitry (not shown) for feeding power at appropriate voltage and current levels to the source 26, the detector 28 and other downhole circuits. These circuits include an amplifier 38 and associated circuitry which receives the output pulses from the PMT 32. The amplified pulses are then applied to a pulse height analyzer (PHA) 40 including an analog-to-digital converter which may be of any conventional type such as the single ramp (Williams readout) type. Other suitable analog to digital converters may be used for the gamma ray energy range to be analyzed. Linear gating circuits may also be employed for control of the time portion of the detector signal frame to be analyzed. Improved performance can be obtained by the use of additional conventional techniques such as pulse pile-up rejection.

The pulse height analyzer 40 assigns each detector pulse to one of a number (typically in the range 256 to 2000) of predetermined channels according to its amplitude (i.e., the gamma ray energy), and produces a signal in suitable digital form representing the channel or amplitude of each analyzed pulse. Typically the pulse height analyzer 40 includes memory in which the occurrences of each channel number in the digital signal are accumulated to provide an energy spectrum. The accumulated totals are then transferred via a buffer memory 42 (which can be replaced in certain circumstances) to telemetry and cable interface circuits 44 for transmission over the cable 15 to the surface equipment 24.

At the surface the cable signals are received by cable interface and signal processing circuits 46. It will be understood that the circuits 44 and 46 may be of any suitable known construction for encoding and decoding, multiplexing and demultiplexing, amplifying and otherwise processing the signals for transmission to and reception by the surface equipment 24. Appropriate circuits are described, for example, in U.S. Pat. No. 4,012,712 to Nelligan.

The operation of the sonde 11 is controlled by signals sent downhole from a master programmer 48, located in the surface equipment 24. These signals are received by a



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tool programmer 50 which transmits control signals to the neutron source 26 and the pulse height analyzer 40.

The surface equipment 34 includes various electronic circuits used to process the data received from the downhole equipment, analyze the energy spectrum of the detected gamma radiation, extract therefrom information about the formation 17 and any hydrocarbons that it may contain, and produce a tangible record or log of some or all of this data and information, for example on film, paper or tape. These circuits may comprise special purpose hardware or alternatively a general purpose computer appropriately programmed to perform the same tasks as such hardware. Details of such analysis form no part of this invention and will not be described here, but may be found for example in U.S. Pat. No. 3,521,064.

Although the invention has been described and illustrated by reference to representative embodiments thereof, it will be understood that such embodiments

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are susceptible of modification and variation without departing from the inventive concepts disclosed. All such modifications and variations, therefore, are intended to be included within the spirit and scope of the appended claims.

I claim:

1. A gamma ray or x-ray detector, comprising a scintillator composed of a transparent single crystal of cerium-activated lanthanum oxyorthosilicate having the general formula  $\text{Ce}_x\text{La}_{1-x}\text{SiO}_4$ , where  $x$  is within the range of from approximately  $2 \times 10^{-4}$  to approximately  $3 \times 10^{-2}$ , and

a photodetector optically coupled to the scintillator for producing an electrical signal in response to the emission of a light pulse by the scintillator.

2. The detector of claim 1 wherein  $x$  is within the range of approximately  $1 \times 10^{-3}$  to approximately  $4.5 \times 10^{-3}$ .

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PATENT APPLICATION SERIAL NO. \_\_\_\_\_

U.S. DEPARTMENT OF COMMERCE  
PATENT AND TRADEMARK OFFICE  
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RECEIVED 02/04/09 0389502 02-4377 010 101 0.00CH

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LOTETIUM ORTHOSILICATE SINGLE  
CRYSTAL SCINTILLATOR DETECTOR

ABSTRACT

A scintillator for use as a gamma ray or like radiation detector is composed of a single crystal of cerium-activated lutetium oxyorthosilicate having the general formulation  $\text{Ce}_{2x}\text{Lu}_{2(1-x)}\text{SiO}_5$ . In a borehole logging application, the detector is mounted in a logging sonde with a high energy neutron source, for movement through a borehole traversing earth formations. Gamma radiation from the surrounding formations is detected and analyzed to provide information concerning hydrocarbons in the formations.

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DESCRIPTION

LUTETIUM ORTHOSILICATE SINGLE  
CRYSTAL SCINTILLATOR DETECTOR

BACKGROUND OF THE INVENTION

This application is a continuation of Application Serial Number 254,353, filed October 6, 1989. *now abandoned*

The present invention relates to a single crystal scintillation detector for gamma rays and like radiation and, more particularly, to a single crystal scintillation detector composed of lutetium orthosilicate.

A well-known form of detector for gamma rays and like radiation (such as x-rays, cosmic rays, and energetic particles of approximately 1 KeV and above) employs a transparent single crystal, known as a scintillator, which responds to impinging radiation to emit light pulses. The light pulses are optically coupled to the input of a photomultiplier tube, which generates a voltage signal related to the number and amplitude of the light pulses received. Scintillators of this class have found wide application in various fields, such as nuclear medicine, physics, chemistry, mineral and petroleum exploration, etc.

Perhaps the most widely used type of scintillator is thallium-doped sodium iodide, NaI (Tl). Relatively inexpensive to produce and capable of providing a high light output in response to impinging radiation, NaI detectors have found general use, for example, in logging tools for oil well logging operations, where either naturally occurring or induced gamma radiation is detected to aid in the location of petroleum deposits.

Other known single crystal scintillators used for gamma ray detection include cesium iodide (sodium or thallium activated) and bismuth germanate (BGO). Or-

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ganic scintillators, such as naphthalene, anthracene, stilbene and similar materials, have also been employed, particularly where very high count rates are important, although they generally are not as useful as inorganic scintillators for the detection of gamma rays.

All of the foregoing types of scintillators have one or more disadvantages as gamma ray detectors. For example, NaI scintillators have comparatively low density, and thus low radiation detection efficiency, slow scintillation decay and a large and persistent afterglow, which impair counting rate performance and lead to pulse pile-up, and are hygroscopic. Although BGO scintillators do not suffer from the low density and hygroscopicity problems of NaI scintillators, they do have a relatively slow scintillation decay time and low light output, which drops still lower at higher temperatures. The index of refraction of BGO is also relatively high, resulting in light loss by internal reflection. These and other disadvantages of known scintillators have limited their usefulness as gamma ray detectors or have imposed restrictions on the manner of their use. In the harsh conditions (high temperature, high pressure, moisture, etc.) of oil well logging, for example, restrictions in logging speed, statistical reliability, tool size and the like have resulted directly from the scintillator material employed in a given logging tool. Accordingly, efforts have long continued to develop still more useful and reliable scintillation detectors and gamma ray scintillation detectors in particular.

More recently, a gamma ray detector employing a scintillator formed of a single crystal of cerium-activated gadolinium orthosilicate (GSO) has been proposed. The GSO scintillator has the advantages as a gamma ray detector of high effective atomic number, high density, fast scintillation decay, relatively low index of refraction, but has the disadvantages of low light

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output, a strong tendency to cleave which makes cutting and polishing difficult, and, more significantly, very high thermal neutron capture cross section (49,000 barns). This last characteristic strongly suggested that GSO scintillators would have very limited utility, if any, in those applications, such as many nuclear well logging tools for instance, where the gamma radiation to be detected is induced by neutron irradiation. This is because gadolinium, upon the capture of thermal neutrons, emits gamma radiation which would interfere with the detection of the external gamma rays of interest.

Such a GSO scintillator detector is described in U. S. Patent No. 4,647,781, issued March 3, 1987, for use in positron computed tomography. There is no disclosure in the '781 patent, however, of the suitability of the GSO scintillator as a gamma ray detector in the hostile conditions of borehole logging. Indeed, it was the present inventor and his co-workers who, through experimentation and evaluation, first discerned that the GSO scintillator was useful as a borehole detector. This invention is disclosed in the copending, commonly-owned U. S. application, Serial No. 149,953 filed February 2, 1988, by Charles H. Reicher et al., which is a continuation of U. S. application Serial No. 812,220 filed December 23, 1985.

Success in adapting the GSO scintillator to borehole use led the inventor to consider other rare earth compounds as possible scintillators for gamma ray (and the like) detection. As a first step, various phosphor materials were studied in powder form for initial evaluation in order to identify potential candidates for crystal growth. This initial evaluation was done using procedures similar to those described by A. H. Gomes de Mesquita et al. at pages 643-650 of Materials Research Bulletin, Vol. 4, No. 9, 1969, Pergamon Press, Inc., who were investigating the properties of rare-earth phos-

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phors as their primary goal. The term phosphor as used in the Gomes de Mesquita et al. paper and as used herein refers to the fine powdered form. Such phosphors are typically used as coatings on cathode ray tube screens, fluorescent light bulbs, and the like, where they convert impinging electrons or ultraviolet radiation into visible light pulses. They are, however, not suitable as gamma ray or like radiation detectors since energetic photons or particles have high probability of passing through the thin coating with no interaction. If the coating is made sufficiently thick to stop gamma rays, the resulting opacity of the phosphor layer would trap most of the scintillation signal. Consequently, only transparent single crystals are useful as gamma ray detectors.

Although some of the physical and optical properties of the rare earth phosphors were known, their scintillation properties were not known and could not be predicted in advance because of the complex and only partially understood nature of the scintillation mechanism. For example, on the basis of atomic number, ionic radius, electronic charge, density, refractive index, and absence of absorption bands, cerium-doped ytterbium orthosilicate powder and cerium-doped lutetium orthosilicate powder would be expected to have very similar scintillation properties. In fact, however, the phosphor experiments showed that ytterbium orthosilicate has 1000 times less scintillation output compared to lutetium orthosilicate.

Moreover, it was not at all clear whether the luminescence properties or other properties of the single crystal form of a rare earth compound would be the same as those of the phosphor form. Generally, in fact, the scintillation properties of single crystal scintillators are not readily determinable or predictable with certainty in advance from the phosphors.

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SUMMARY

In accordance with the present invention, therefore, an improved scintillator for use as a gamma ray (or like radiation) detector is provided which comprises a single crystal of cerium-activated lutetium oxyorthosilicate having the general formulation  $\text{Ce}_x\text{Lu}_{2(1-x)}\text{SiO}_5$ . Generally, the value of  $x$  (as measured in the initial melt from which the crystal is pulled) may be varied within the approximate range of from 0.001 to 0.1, with the preferred range of  $x$  being from approximately 0.005 to 0.015. When assembled in a complete detector, the scintillator crystal is optically coupled, either directly or through a suitable light path, to the photosensitive surface of a photodetector for generation of an electrical signal in response to the emission of a light pulse by the scintillator. The LSO scintillator of the invention possesses certain important characteristics, most notably high light output, very short decay time and high detection efficiency, that make it superior to prior scintillators as a gamma ray or like radiation detector, particularly in the borehole logging environment.

BRIEF DESCRIPTION OF THE DRAWING

Figure 1 is a diagrammatic view of one embodiment of an LSO scintillation detector in accordance with the present invention; and

Figure 2 is a schematic diagram of a borehole logging sonde in which the LSO scintillation detector of Figure 1 may be used.

DESCRIPTION OF REPRESENTATIVE EMBODIMENT

For illustrative purposes, a representative embodiment of the invention is described hereinafter in the context of a gamma ray detector. It will be understood, of course, that the utility of the novel LSO single

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crystal scintillator of the invention is not limited to the detection of gamma radiation but that it has general application to the detection of other types of like radiation, e.g. X-rays, cosmic rays, and energetic particles.

In Fig. 1, a single crystal LSO scintillator 10 is shown encased within the housing 12 of a gamma ray detector. One face 14 of the scintillator is placed in optical contact with the photosensitive surface of a photomultiplier tube 16. Alternatively, the light pulses could be coupled to the photomultiplier via light guides or fibers, lenses, mirrors, or the like. The photomultiplier can be replaced by any suitable photodetector such as a photodiode, microchannel plate, etc. In order to direct as much of each light flash to the photomultiplier as possible, the other faces 18 of the scintillator are preferably surrounded or covered with a reflective material, e.g. Teflon tape, magnesium oxide powder, aluminum foil, or titanium dioxide paint. Light pulses emitted by the LSO crystal upon the incidence of radiation are intercepted, either directly or upon reflection from the surfaces 18, by the photomultiplier, which generates electrical pulses or signals in response to the light pulses. These electrical output pulses are typically first amplified and then subsequently processed as desired, e.g. in a pulse height amplifier, to obtain the parameters of interest regarding the detected radiation. The photomultiplier is also connected to a high voltage power supply, as indicated in Fig. 1. Other than the LSO scintillator, all of the components and materials referred to in connection with Fig. 1 are conventional, and thus need not be described in detail.

As the first step in the evaluation of cerium-activated lutetium oxyorthosilicate  $\text{Ce:Lu}_2\text{SiO}_5$  as a scintillator, the material was synthesized in phosphor

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(powder) form and the scintillation properties of the resulting compound were analyzed. From this analysis, it was determined that LSO was sufficiently promising as a scintillator material to warrant the additional effort and expense of growing in single crystal form. LSO was chosen as a candidate for phosphor synthesis because of its high atomic number and high density and the absence of optical absorption bands in the visible part of the spectrum. The principal physical and optical properties of LSO are compared to those of GSO and NaI (Tl) in Table 1, from which it may be seen that LSO appears to offer advantages over GSO in all properties and over NaI (Tl) in the areas of atomic number, density, index of refraction and hygroscopicity.

TABLE 1

	NaI(Tl)	GSO	LSO
Effective atomic number	51	59	66
Density (gm/cm <sup>3</sup> )	3.67	6.71	7.4
Index of Refraction	1.85	1.91	1.82
Neutron cross section (barns)	6.2	49,000	84
Hygroscopic?	yes	no	no

As Ce-activated single crystals of LSO had not previously been grown, the scintillation properties of single-crystal LSO were unknown. Also unknown, therefore, was whether the properties of the LSO phosphor would be retained when the same material was grown into a single crystal.

The phosphor synthesis procedure consisted of the following steps:

- (1) Fine powders of Ln<sub>2</sub>O<sub>3</sub> (Ln=Gd, Lu) SiO<sub>2</sub> and either CeO<sub>2</sub> or Ce<sub>2</sub>O<sub>3</sub> in the appropriate atomic ratios were mechanically mixed and loaded into an alumina boat. In each case the molar concentration of Ce was 0.5% relative to Gd or Lu.

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(2) Each of the four compositions was sintered at 1500°C for four hours in a tube oven in one of three atmospheres: neutral (pure argon), reducing (argon + 2% hydrogen), or oxidizing (air). When a reducing atmosphere was used, a high purity alumina furnace tube was employed because the standard mullite tube was found to react with hydrogen at high temperature and contaminate the samples. The resulting material was solid and was ground into a powder for analysis.

(3) The fluorescence emission of the powder was excited with ultraviolet light and the emission and excitation spectra were recorded with a Spex Fluorolog (Model 212) spectrofluorometer. For emission measurements above room temperature, the sample chamber was continuously flushed with argon to suppress spurious fluorescence emission which occurred when the sample was heated above 100°C in air.

Table 2 shows the intensity of the fluorescence emission measured at room temperature.

TABLE 2

	GSO:CeO <sub>2</sub>	GSO:Ce <sub>2</sub> O <sub>3</sub>	LSO:CeO <sub>2</sub>	LSO:Ce <sub>2</sub> O <sub>3</sub>
Argon + H <sub>2</sub>	44	17	71	27
air	1.8	1.6	11	28
Argon	12	8.5	100	86

(4) The fluorescence decay times of the phosphors were measured by spreading a thin layer of the phosphor on the face of a fast photomultiplier (Amperex 2020Q) and exciting the emission with an <sup>241</sup>Na gamma-ray source. The pulse shape was observed at the anode of the photomultiplier with a fast oscilloscope and the decay time estimated from the oscilloscope trace.

Although the synthesis conditions employed were not necessarily optimum for each phosphor, inasmuch as the emission intensity of LSO was greater than that of GSO under all six synthesis conditions (three different

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atmospheres and two forms of cerium), it may be concluded that LSO's scintillation efficiency is intrinsically greater. The scintillation mechanism in cerium-activated phosphors is generally assumed to involve electronic transitions (5d to 4f) within the  $\text{Ce}^{+3}$  ion. For this reason, it was expected that the most efficient phosphors would result from using  $\text{Ce}_2\text{O}_3$  (i.e.,  $\text{Ce}^{+3}$ ) as a starting material and performing the synthesis in a slightly reducing atmosphere ( $\text{Ar} + \text{H}_2$ ) in order to maintain the cerium in the +3 charge state. Surprisingly, as Table 2 shows, in every case except LSO prepared in air the best phosphors were those in which Ce was added as  $\text{CeO}_2$ . Also note that although the best GSO phosphors were those prepared in a reducing atmosphere, the best LSO phosphors were those prepared in a neutral atmosphere. Synthesis in air resulted in phosphors with lower light output and, more importantly, with emission spectra that differed from the single crystal emission spectrum.

Table 3 summarizes the scintillation properties of GSO and LSO phosphors, synthesized in the foregoing manner:

	<u>TABLE 3</u>	
	GSO: $\text{CeO}_2$	LSO: $\text{CeO}_2$
Light output	1	1.5-10
Decay time	60 ns	50 ns
Emission peak	430 nm	415 nm
Temperature response	-0.4%/°C	-1.3%/°C

A comparison of the physical properties (Table 2) and the scintillation properties (Table 3) of GSO and LSO show LSO to be superior to GSO in most areas. The light output of LSO is a factor of 1.5 to 10 greater than GSO depending on the synthesis conditions. This is a particularly notable advantage over GSO. A comparison of the emission spectra of the GSO: $\text{CeO}_2$  and LSO: $\text{CeO}_2$

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phosphors synthesized in argon (with the excitation light wavelength corresponding to the strongest excitation band for each phosphor, 345 nm for GSO and 355 nm for LSO) shows the LSO emission to have a different shape and to be shifted somewhat towards the shorter wavelengths than the GSO emission. This apparent shift of the LSO spectrum, if exhibited in the single crystal form, would be advantageous since it would result in an emission that better matches the spectral response of high temperature photomultipliers.

The scintillation decay time of LSO of about 50 ns compares favorably with GSO's 60 ns. The effective atomic number of LSO is 66 compared to 59 for GSO and the density of LSO is 7.4 gm/cc compared to 6.7 gm/cc for GSO, both of which contribute to a higher radiation detection efficiency for LSO. The index of refraction of LSO is 1.82 compared to 1.91 for GSO, which results in less trapping of scintillation light. LSO is very much less sensitive to neutrons, since the thermal cross section for Lu is 77 barns compared to 49,000 barns for Gd.

The temperature response of LSO is somewhat worse than GSO. At 150°C, GSO's light output decreases to about 60% of its room temperature value, while LSO's light output decreases to about 20% of its room temperature value at 150°C. On the other hand, the emission spectrum of LSO was found to shift somewhat to shorter wavelengths than the GSO emission as temperature increased from room temperature up to 175°C (the highest temperature measured). Again this would be advantageous in the single crystal form in terms of matching photomultiplier response.

Lutetium has a radioactive isotope ( $^{176}\text{Lu}$ ) that produces a background noise level in the crystal. This could be eliminated by growing the crystal from pure

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$^{175}\text{Lu}$ , or it could be handled by conventional background subtraction techniques.

The excitation spectrum of LSO exhibits three bands (262 nm, 298 nm, and 355 nm) and is similar to the GSO excitation spectrum except that the bands are shifted to somewhat longer wavelengths.

Although the comparison of the LSO and GSO phosphors showed LSO to be a promising scintillator for use in a gamma ray detector, the properties of the single crystal form cannot be predicted with certainty. Light output, one of the most important properties of a single crystal scintillator, is especially difficult to predict from the phosphor form. In order to continue with a more detailed evaluation of the material as well as to test a practical detector, therefore, it was necessary to grow a single crystal form of LSO. This was done from melts by the conventional Czochralski method, as described, for example, by C. D. Brandle et al., "Czochralski Growth of Rare-Earth Orthosilicates ( $\text{Ln}_2\text{SiO}_5$ )", Journal of Crystal Growth, No. 79, pp. 308-315, 1986.

Table 4 summarizes the scintillation properties of the LSO single crystals grown, as selected and cut to minimize imperfections. The composition of the melts was  $\text{Ce}_2\text{xLu}_{2(1-x)}\text{SiO}_5$ , where x is the decimal value of the percentage set out in Table 4 under the heading "Ce nom". Cerium concentration in the crystals was on the order of 20%-30% of that in the melt.

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TABLE 4

	size (mm)	color	defects	Ca nom- (%)	light output (arbitrary unit)	energy res (%)	decay constant (ns)	emission max (UV excitation (nm)	emission max (gamma excitation) (nm)
1	5x5x1	none	none	1.0	156	8.8	44	395	428
2	7x9x1	none	few	1.3	109	12	41	395	430
3	5x5x25	none	none	0.7	146	11.2	44	394	428
4	5x5x1	none	few	0.6	104	13	38	393	426

TABLE 4

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Crystals 1, 2 and 4 were cut to the sizes listed from larger single crystals (5mm x 6mm x 20mm, 7mm x 9mm x 27mm and 6mm x 8mm x 33mm, respectively), but crystal 3 was the original size. All were clear of color and of high transparency, but crystals 2 and 4 had some defects. The light output was measured by coupling each crystal with optical coupling grease directly to a Hamamatsu R878 photomultiplier, with all surfaces except that coupled to the photomultiplier covered with Teflon tape (crystal 3) or titanium dioxide paint (crystals 1, 2 and 4). The scale employed for the light output measurements is expressed in arbitrary units. By comparison, on this scale the light output of a standard NaI (Tl) scintillator would be on the order of 200, and that for a standard GSO scintillator would be on the order of 40. The energy resolution was determined by using a standard cesium 137 gamma ray source. The energy resolution is expressed as the full width at half-maximum of the 662 KeV gamma ray peak. The scintillation decay time was exponential and had an average value of about 42 ns among the four crystals, as measured by the time-correlated, single photon technique.

Unexpectedly, the emission spectrum under gamma excitation was found to be different from the emission spectrum under ultraviolet excitation. The gamma emission spectrum peaked at approximately 426 nm -430 nm and was similar to the GSO emission spectrum. Also, both the gamma and ultraviolet-excited emissions exhibited a thermoluminescent effect with a half-life of about 10 minutes. The temperature response of the LSO crystals was not as good as GSO. The gamma-excited emission fell off at approximately 1.3% per degree C. Thus at 150°, the peak output is 20% of that at room temperature, which is similar to the temperature response of BGO. Accordingly, in those applications where high temperatures are anticipated, such as in certain oil well log-

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ging tools, the LSO scintillator may need to be isolated from the environment by a Dewar flask or other insulator.

Based on the data of Table 4 and previous experience with other single crystal scintillators and powder phosphors, it is expected that satisfactory LSO single crystal scintillators can be produced with cerium concentrations (in the melt from which the crystal is pulled) within the approximate range of from 0.1% to 10%, i.e.,  $0.001 \leq x \leq 0.1$ . The preferred melt cerium concentration is within the range of from approximately 0.5% to 1.5%, i.e.,  $0.005 \leq x \leq 0.015$ .

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TABLE 5

	NaI(Tl)	BGO	GSO	LSO
Relative light output (20°C)	100	12	20	75
Energy resolution (best)	6%	9%	8%	9%
				-- signal-to-noise
Decay time (ns)	230	300	60	41
				-- counting rate
Effective atomic no.	51	75	59	66
Density (gm/cm <sup>3</sup> )	3.67	7.13	6.71	7.4
				-- detection eff.
Index of refraction	1.85	2.15	1.91	1.82
Hygroscopic?	yes	no	no	no
Mechanically rugged?	no	yes	no	yes
				-- packaging
Emission peak (nm at 20°C)	410	480	430	428
neutron cross section (barns)	6.2	2.2	49,000	84
radiation length (cm)	2.6	1.1	1.4	1.1

TABLE 5

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Table 5 compares the principal physical and scintillation properties of the LSO single crystals with those for NaI(Tl), BGO, and GSO. With the NaI(Tl) crystal arbitrarily assigned a reference light output value of 100, it may be seen that the LSO crystal at 75 is markedly superior to the BGO and GSO crystals and only 25% below the NaI(Tl) value. The energy resolution of the LSO scintillator compares quite favorably with BGO and GSO and, again, was only slightly worse than NaI(Tl). The signal-to-noise performance of the LSO scintillator, therefore, is much improved relative to the BGO and GSO detectors. Although somewhat lower than NaI(Tl) in signal-to-noise ratio, LSO possesses other properties that are superior to NaI(Tl).

Thus the average decay time of 41 ns is shorter than any of the other three crystals and is some 5 to 6 times shorter than NaI(Tl). The LSO scintillator, therefore, is particularly useful in high counting rate detectors. LSO also has a very high gamma ray detection efficiency by virtue of its high effective atomic number and density. It is superior in this respect to both NaI(Tl) and GSO and is comparable to BGO. High detection efficiency further contributes to LSO's suitability for high counting rate applications.

Other favorable properties of LSO apparent from Table 5 include its low index of refraction, which results in less internal light loss than any of the other four scintillators. LSO is also non-hygroscopic, a particular advantage for oil well logging applications or other wet environments. Its mechanical ruggedness is superior to both NaI(Tl) and GSO, a feature which is also desirable for well logging and other uses where rough handling is encountered.

As already noted, the gamma emission peak is at approximately 428 nm, which is substantially the same as GSO and only slightly above NaI(Tl). A good match to

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the typical photomultiplier spectral response results. The neutron cross section is especially favorable in comparison to GSO, 84 barns vs. 49,000 barns. Hence the occurrence of interfering gamma rays due to neutron capture within the crystal is greatly reduced relative to GSO. Finally, the radiation length of LSO is as good as that of BGO and considerably shorter than either GSO or NaI, with consequent advantages in the crystal size required.

As may be appreciated from Tables 4 and 5, the LSO single crystal scintillator is comparable to or exceeds other known scintillators in nearly all properties important for use as a gamma ray detector, i.e., light output, energy resolution, efficiency of detection of high energy photons, scintillation decay time, hygroscopicity, susceptibility of crystal to mechanical damage, refractive index, emission spectrum match to photomultiplier tube response, transparency of the crystal to its own scintillation emission, and absence of induced gamma radiation within the crystal. The only area in which LSO compares unfavorably is in the temperature sensitivity of the gamma-excited emission. In controlled environments, e.g. laboratories, hospitals, etc., this presents no problem. Even in high temperature environments, moreover, the unique combination of light output, speed, and detection efficiency found in LSO will in many instances justify the additional effort of isolating the scintillator from environmental temperature changes.

As discussed above, the LSO scintillator detector of the present invention is particularly effective as a radiation detector in a borehole logging environment, such as for petroleum exploration. In such use, the detector forms part of a logging system which may be of the type disclosed in the aforementioned copending

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application Serial No. 149,953 and illustrated in Figure 2 herein.

Figure 2 shows a logging sonde 11 for sensing gamma radiation resulting from bombardment of a formation with high energy neutrons and detecting the energy of the radiation for subsequent spectral analysis. The sonde 11 is suspended in a borehole 13 on an armored multi-conductor cable 15. The borehole 13 traverses a formation 17 and is filled with fluid 19, and may be open as shown or cased. The sonde 11 as described below may be constructed in accordance with U.S. Patent No. 4,317,993 to Bertzog, Jr. et al, assigned to the assignee of the present application. The sonde 11 is moved in the borehole 13 by paying the cable 15 out and reeling it back in over a sheave wheel 20 and a depth gauge 22 by means of a winch forming part of a surface equipment 24. Usually the logging measurements are actually made while the sonde 11 is being raised back up the borehole 13, although in certain circumstances they may be made on the way down instead or as well.

The sonde 11 includes a pulsed neutron source 26 for producing primary radiation to bombard the formation 17 with fast neutrons as the sonde 11 travels up the borehole 13, and a radiation detector 28 for detecting secondary (gamma) radiation induced thereby in the borehole 13 and the formation 17. The neutron source 26 is preferably of the pulsed accelerator type described in U.S. Patents No. 3,461,291 to Goodman and No. 3,546,512 to Prentrop, both commonly owned with this application. This type of source is particularly suited to the generation of discrete bursts of high energy or fast neutrons, e.g. at 14 MeV, with a controlled duration and repetition rate.

The detector 28 is of a type appropriate to the detection of gamma radiation and the production of an electrical signal corresponding to each detected gamma

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ray and having an amplitude representative of the energy of the gamma ray. To this end the detector 28 is as shown in Figure 1, including a cerium-activated LSO scintillation crystal 10 optically coupled to a photomultiplier tube (PMT) 16. Suitable tubes are manufactured by EMR Photoelectric, Princeton, New Jersey.

Although not as important as in the case of a GSO scintillator, a neutron shield 34 may be located between the source 26 and the detector 28 to limit direct bombardment of the detector 28 by neutrons from the source 26, thereby avoiding saturation of the detector 28 by such direct irradiation. In addition, especially in the case of measurement of capture gamma radiation, the sonde 11 may be surrounded by a sleeve 36 impregnated with boron carbide and located in the general vicinity of the source 26 and the detector 28. This sleeve displaces borehole fluid in the region of the detector 28, and absorbs neutrons scattered by the formation towards the detector 28, without significantly attenuating gamma radiation emanating from the formation. The net effect is to reduce the possibility of neutron interactions with the borehole contents and the material of the sonde 11 in proximity to the detector 28 and which would otherwise produce detectable gamma rays constituting an undesirable perturbation of the required gamma ray measurement.

Electrical power for the sonde 11 is supplied via the cable 15 from the surface equipment 24. The sonde 11 includes power conditioning circuitry (not shown) for feeding power at appropriate voltage and current levels to the source 26, the detector 28 and other downhole circuits. These circuits include an amplifier 38 and associated circuitry which receives the output pulses from the PMT 16. The amplified pulses are then applied to a pulse height analyzer (PHA) 40 including an

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analog-to-digital converter which may be of any conventional type such as the single ramp (Wilkinson rundown) type. Other suitable analog to digital converters may be used for the gamma ray energy range to be analyzed. Linear gating circuits may also be employed for control of the time portion of the detector signal frame to be analyzed. Improved performance can be obtained by the use of additional conventional techniques such as pulse pile-up rejection.

The pulse height analyzer 40 assigns each detector pulse to one of a number (typically in the range 256 to 8000) of predetermined channels according to its amplitude (i.e. the gamma ray energy), and produces a signal in suitable digital form representing the channel or amplitude of each analyzed pulse. Typically the pulse height analyzer 40 includes memory in which the occurrences of each channel number in the digital signal are accumulated to provide an energy spectrum. The accumulated totals are then transferred via a buffer memory 42 (which can be omitted in certain circumstances) to telemetry and cable interface circuits 44 for transmission over the cable 15 to the surface equipment 24.

At the surface the cable signals are received by cable interface and signal processing circuits 46. It will be understood that the circuits 44 and 46 may be of any suitable known construction for encoding and decoding, multiplexing and demultiplexing, amplifying and otherwise processing the signals for transmission to and reception by the surface equipment 24. Appropriate circuits are described, for example, in U.S. Patent No. 4,012,712 to Nelligan.

The operation of the sonde 11 is controlled by signals sent downhole from a master programmer 48, located in the surface equipment 24. These signals are received by a tool programmer 50 which transmits control

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signals to the neutron source 26 and the pulse height analyzer 40.

The surface equipment 24 includes various electronic circuits used to process the data received from the downhole equipment, analyze the energy spectrum of the detected gamma radiation, extract therefrom information about the formation 17 and any hydrocarbons that it may contain, and produce a tangible record or log of some or all of this data and information, for example on film, paper or tape. These circuits may comprise special purpose hardware or alternatively a general purpose computer appropriately programmed to perform the same tasks as such hardware. Details of such analysis form no part of this invention and will not be described here, but may be found for example in U.S. Patent No. 3,521,064.

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Although the invention has been described and illustrated by reference to representative embodiments thereof, it will be understood that such embodiments are susceptible of modification and variation without departing from the inventive concepts disclosed. All such modifications and variations, therefore, are intended to be included within the spirit and scope of the appended claims.

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CLAIMS

1. A scintillator for use in a gamma ray or x-ray detector, comprising a transparent single crystal of cerium-activated lutetium oxyorthosilicate having the general formulation  $\text{Ce}_{2x}\text{Lu}_{2(1-x)}\text{SiO}_5$ , where x is within the range of from approximately  $2 \times 10^{-4}$  to approximately  $3 \times 10^{-2}$ .
2. The scintillator of claim 1 wherein x is within the range of approximately  $1 \times 10^{-3}$  to approximately  $4.5 \times 10^{-3}$ .
3. A gamma ray or x-ray detector, comprising: a scintillator composed of a transparent single crystal of cerium-activated lutetium oxyorthosilicate having the general formulation  $\text{Ce}_{2x}\text{Lu}_{2(1-x)}\text{SiO}_5$ , where x is within the range of from approximately  $2 \times 10^{-4}$  to approximately  $3 \times 10^{-2}$ , and  
a photodetector optically coupled to the scintillator for producing an electrical signal in response to the emission of a light pulse by the scintillator.
4. The detector of claim 3 wherein x is within the range of approximately  $1 \times 10^{-3}$  to approximately  $4.5 \times 10^{-3}$ .
5. Apparatus for investigation of subsurface earth formations comprising:
  - a sonde adapted for movement through a borehole;
  - detector means carried by said sonde for detecting radiation and including a lutetium oxyorthosilicate scintillator; and
  - means coupled to said detector means for generating and recording a signal representative of at least one characteristic of radiation detected by said detector means.

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-23-

6. The apparatus of claim 5 above wherein said scintillator is composed of a single transparent crystal of cerium-activated lutetium oxyorthosilicate having the general formulation  $\text{Ce}_{2x}\text{Lu}_{2(1-x)}\text{SiO}_5$ , where  $x$  is within the range of from approximately  $2 \times 10^{-4}$  to approximately  $3 \times 10^{-2}$ .
7. The apparatus of claim 5, above wherein said scintillator is composed of a single transparent crystal of cerium-activated lutetium oxyorthosilicate having the general formulation  $\text{Ce}_{2x}\text{Lu}_{2(1-x)}\text{SiO}_5$ , wherein  $x$  is within the range of from approximately  $2 \times 10^{-4}$  to approximately  $3 \times 10^{-2}$ , and wherein said detector means further comprises
  - a photodetector optically coupled to the scintillator for producing an electrical signal in response to the emission of a light pulse by the scintillator.
8. The apparatus of claims 6 or 7 above wherein  $x$  is within the range of approximately  $1 \times 10^{-3}$  to approximately  $4.5 \times 10^{-3}$ .
9. Apparatus for investigation of subsurface earth formations comprising:
  - a sonde adapted for movement through a borehole;
  - source means carried by said sonde for irradiating material in the region of said borehole with penetrating radiation capable of interaction with said material resulting in radiation having characteristics carrying information about said material;
  - detector means carried by said sonde radiation and including a lutetium oxyorthosilicate scintillator; and

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means coupled to said detector means for generating and recording a signal representative of at least one characteristic of radiation detected by said detector means.

10. The apparatus of claim 9 above wherein said scintillator is composed of a single transparent crystal of cerium-activated lutetium oxyorthosilicate having a general formulation  $\text{Ce}_{2x}\text{Lu}_{2(1-x)}\text{SiO}_5$ , where  $x$  is within the range of from approximately  $2 \times 10^{-4}$  to approximately  $3 \times 10^{-2}$ .
11. The apparatus of claim 9, above wherein said scintillator is composed of a single transparent crystal of cerium-activated lutetium oxyorthosilicate having the general formulation  $\text{Ce}_{2x}\text{Lu}_{2(1-x)}\text{SiO}_5$ , where  $x$  is within the range of from approximately  $2 \times 10^{-4}$  to approximately  $3 \times 10^{-2}$ , and wherein said detector means further comprises  
 a photodetector optically coupled to the scintillator for producing an electrical signal in response to the emission of a light pulse by the scintillator.
12. The apparatus of claims 9 or 10 above wherein  $x$  is within the range of approximately  $1 \times 10^{-3}$  to approximately  $4.5 \times 10^{-3}$ .

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PATENT  
25715-I 539/15132

# DECLARATION FOR PATENT APPLICATION AND POWER OF ATTORNEY

As a below named inventor, I hereby declare that:

My residence, post office address(es) and citizenship(s) are as stated below next to my name, and

I believe I am the original, first and sole inventor of the subject matter which is claimed (if only one name is listed below) or an original and first inventor of at least some of the subject matter which is claimed (if plural names are listed below) and for which a patent is sought on the invention entitled

"Lutetium Orthosilicate Single Crystal Scintillator Detector"

the specification of which

☒ is attached hereto.

☐ was filed on

as Application Serial Number

and was amended on (if applicable).

I hereby state that I have reviewed and understand the contents of the above identified specification, including the claims, as amended by any amendment referred to above.

I acknowledge the duty to disclose information which is material to the examination of this application in accordance with Title 37, Code of Federal Regulations, Section 1.56(a).

## Prior Foreign Application(s)

I hereby claim foreign priority benefits under Title 35, United States Code, Section 119 of any foreign application(s) for patent or inventor's certificate listed below and have also identified below any foreign application(s) for patent or inventor's certificate having a filing date before that of the application on which priority is claimed:

Country	Application No.	Filed(d/m/y)	Issued(d/m/y)	Priority Claimed
				Y <input type="checkbox"/> N <input type="checkbox"/>
				Y <input type="checkbox"/> N <input type="checkbox"/>
				Y <input type="checkbox"/> N <input type="checkbox"/>

## Prior United States Applications

I hereby claim the benefit under Title 35, United States Code, Section 120 of any United States application(s) listed below and, insofar as the subject matter of each of the claims of this application is not disclosed in the prior United States application in the manner provided by the first paragraph of Title 35, United States Code, Section 112, I acknowledge the duty to disclose material information as defined in Title 37, Code of Federal Regulations, Section 1.56(a) which occurred between the filing date of the prior application and the national or PCT international filing date of this application:

Application Serial No.	Filing Date(d/m/y)	Status (Patented, Pending, Abandoned)
254,353	6/10/88	pending

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PATENT  
25715-1 539/15132  
PAGE 2 of 2

And, I hereby appoint, both jointly and severally, as my attorney(s) and/or agent(s) with full power of substitution and revocation, to prosecute this application and to conduct all business in the Patent and Trademark Office connected herewith the following attorney(s) and agent(s), their registration numbers being listed after their names:

John E. Dumarsq, Reg. No. 14,638; Dana M. Raymond, Reg. No. 18,540; Richard G. Fuller, Jr., Reg. No. 18,284; Frank W. Ford, Jr., Reg. No. 16,614; Frederick C. Carver, Reg. No. 17,021; George W. Whitney, Reg. No. 17,099; Allen G. Weiss, Reg. No. 18,021; Francis J. Hunt, Reg. No. 18,662; William P. Eberle, Reg. No. 18,133; Joseph D. Garon, Reg. No. 20,420; Arthur S. Tenner, Reg. No. 18,832; Ronald B. Hildreth, Reg. No. 19,498; Allan H. Bonnell, Reg. No. 22,059; Thomas R. Neublu, Jr., Reg. No. 22,875; Robert Neuner, Reg. No. 24,218; Richard G. Berkley, Reg. No. 25,465; Edward V. Filardi, Reg. No. 25,757; Richard S. Clark, Reg. No. 26,154; Thomas D. MacBlain, Reg. No. 24,583; Bradley B. Geist, Reg. No. 27,551; James A. Malone, Reg. No. 26,946; Robert D. Smith, Reg. No. 28,538; and M. Andyt Ryan, Reg. No. 28,469, of the firm of BRUMBAUGH, GRAVES, DONOHUE & RAYMOND, with offices at 30 Rockefeller Plaza, New York, New York 10112 and Kenneth Olsen, Reg. No. 26,493; Keith G. W. Smith, Reg. No. 32,436; Peter Y. Lee, Reg. No. 30,865; and Clifford L. Tager, Reg. No. 31,704.

I hereby request that all correspondence be directed to BRUMBAUGH, GRAVES, DONOHUE & RAYMOND, 30 Rockefeller Plaza, New York, New York 10112 and that all telephone calls be directed to (212) 403-2500. I hereby declare that all statements made herein of my own knowledge are true and that all statements made on information and belief are believed to be true; and further that these statements were made with the knowledge that willful false statements and the like so made are punishable by fine or imprisonment, or both, under Section 1001 of Title 18 of the United States Code and that such willful false statements may jeopardize the validity of the application or any patent issuing thereon.

#### SOLE OR FIRST INVENTOR

Signature: Charles L. Melcher Date: August 2, 1989

Full Name: Charles L. Melcher

Residence: 49 Lamport Road, West Redding, Connecticut 06896

Citizenship: United States

Post Office Address: 49 Lamport Road, West Redding, Connecticut 06896

#### SECOND JOINT INVENTOR (IF ANY)

Signature: \_\_\_\_\_ Date: \_\_\_\_\_

Full Name: \_\_\_\_\_

Residence: \_\_\_\_\_

Citizenship: \_\_\_\_\_

Post Office Address: \_\_\_\_\_

#### THIRD JOINT INVENTOR (IF ANY)

Signature: \_\_\_\_\_ Date: \_\_\_\_\_

Full Name: \_\_\_\_\_

Residence: \_\_\_\_\_

Citizenship: \_\_\_\_\_

Post Office Address: \_\_\_\_\_

SMS003187

Print of Drawing  
As Original Filed

07/289502

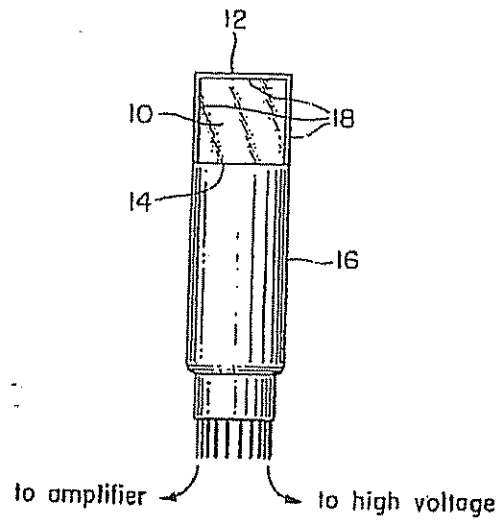
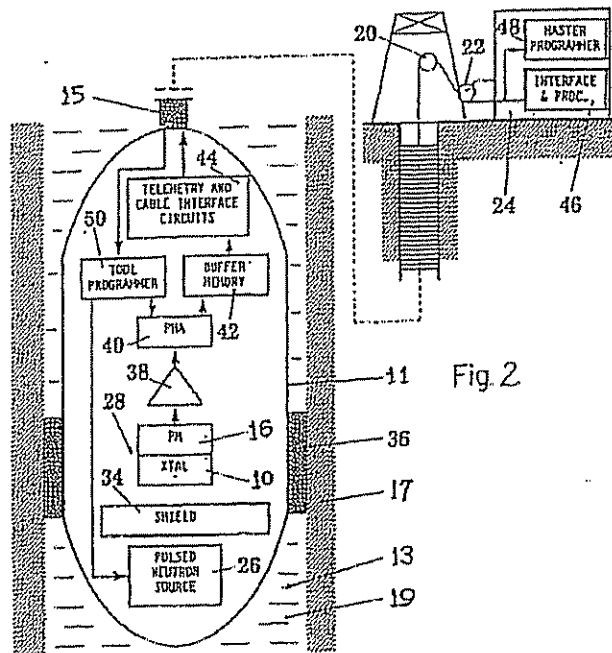


FIG. 1

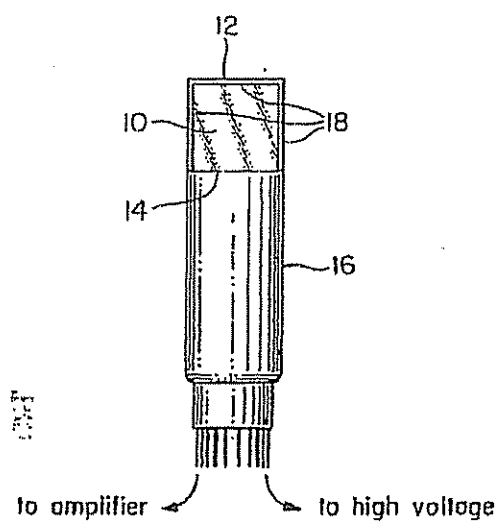
SMS003188

Print of Drawing  
As Original Filed

W/39502

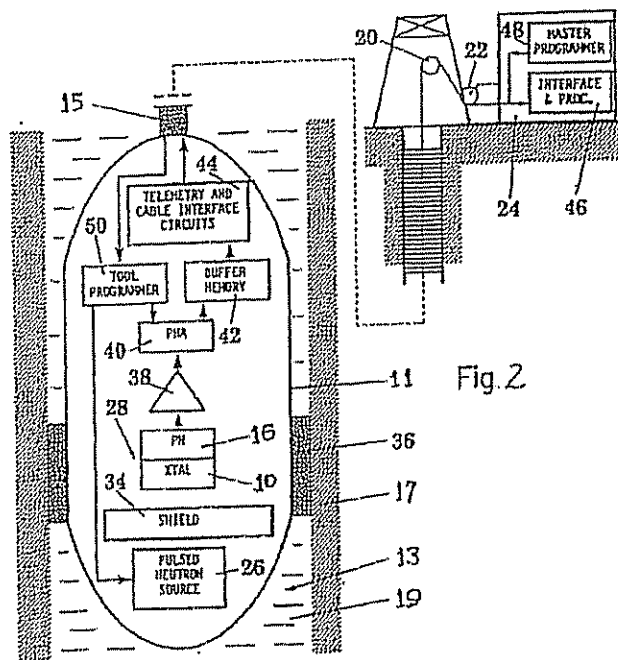


11/389502



SMS003190

07/389502



SMS003191



FORM PTO-875 REV. 1-84	U.S. DEPARTMENT OF COMMERCE PATENT AND TRADEMARK OFFICE	SERIAL NO. 389502	FILING DATE 8/4/89
PATENT APPLICATION FEE DETERMINATION RECORD		APPLICANT'S FIRST NAME MELCHER, L.R.	

## CLAIMS AS FILED - PART I

FOR	NO FILED	NO EXTRA
BASIC FEE		
TOTAL CLAIMS	14 - 20	
INDEP. CLAIMS	4 - 20	1
MULTIPLE DEPENDENT CLAIMS PRESENT		

\* If the application is filed in the form of a divisional application, the fee for the basic fee is \$100.

## SMALL ENTITY

RATE	FEE
	\$ 185
X 6	
X 18	
60	
TOTAL	

OTHER THAN A  
SMALL ENTITY

RATE	FEE
	\$ 370
X 12	
X 36	
120	
TOTAL	\$ 526

## CLAIMS AS AMENDED - PART II

AMENDMENT A	CLAIMS REMAINING AFTER AMENDMENT	HIGHEST NO. PREVIOUSLY PAID FOR	PRESENT EXTRA
	TOTAL	MINUS	
	INDEP.	MINUS	
FIRST PRESENTATION OF MULTIPLE DEP. CLAIM			

## SMALL ENTITY

RATE	ADDIT. FEE
X 6	
X 18	
60	
TOTAL	

OTHER THAN A  
SMALL ENTITY

RATE	ADDIT. FEE
X 12	
X 36	
120	
TOTAL	

AMENDMENT B	CLAIMS REMAINING AFTER AMENDMENT	HIGHEST NO. PREVIOUSLY PAID FOR	PRESENT EXTRA
	TOTAL	MINUS	
	INDEP.	MINUS	
FIRST PRESENTATION OF MULTIPLE DEP. CLAIM			

## SMALL ENTITY

RATE	ADDIT. FEE
X 6	
X 18	
60	
TOTAL	

OTHER THAN A  
SMALL ENTITY

RATE	ADDIT. FEE
X 12	
X 36	
120	
TOTAL	

AMENDMENT C	CLAIMS REMAINING AFTER AMENDMENT	HIGHEST NO. PREVIOUSLY PAID FOR	PRESENT EXTRA
	TOTAL	MINUS	
	INDEP.	MINUS	
FIRST PRESENTATION OF MULTIPLE DEP. CLAIM			

## SMALL ENTITY

RATE	ADDIT. FEE
X 6	
X 18	
60	
TOTAL	

OTHER THAN A  
SMALL ENTITY

RATE	ADDIT. FEE
X 12	
X 36	
120	
TOTAL	

\* If the application is filed in the form of a divisional application, the fee for the basic fee is \$100.  
 \*\* If the application is filed in the form of a divisional application, the fee for the basic fee is \$100.  
 \*\*\* If the application is filed in the form of a divisional application, the fee for the basic fee is \$100.

SMS003192

[illegible]

SMS003193

FORMAT NO. 3 ATTORNEY'S REGISTRATION NO.(S)									
RECORD 301					No. 1				
No. 2					No. 2				
No. 4					No. 5				
No. 6					No. 7				
RECORD 302					No. 1				
No. 2					No. 2				
No. 4					No. 5				
No. 6					No. 7				
RECORD 303					No. 1				
No. 2					No. 2				
No. 4					No. 5				
No. 6					No. 7				
<input type="checkbox"/> MORE ON SUPPLEMENTAL CODING SHEET									

FORMAT NO. 4 APPLICANT(S) NAME AND ADDRESS									
RECORD		AUTHORITY		FAMILY NAME (Last)		NAME-SUFFIX			
4	0	1							
GIVEN NAMES (First)									
CITY (If Applicable)									
STATE/COUNTRY									
RECORD		AUTHORITY		FAMILY NAME (Last)		NAME-SUFFIX			
4	0	2							
GIVEN NAMES (First)									
CITY (If Applicable)									
STATE/COUNTRY									
<input type="checkbox"/> MORE ON SUPPLEMENTAL CODING SHEET									

FORMAT NO. 5 TITLE OF INVENTION (If Special)									
RECORD 501									
RECORD 502									
RECORD 503									
RECORD 504									

FORMAT NO. 6 CORRESPONDENCE ADDRESS (If Special)									
RECORD 601									
RECORD 602									

FORMAT NO. 7									
RECORD 701									
RECORD 702									
RECORD 703									

SMS003194

MULTIPLE DEPENDENT CLAIM PER CALCULATION SHEET (FOR USE WITH FORM PTO-875)							SERIAL # 39502	FILED DATE 8/4/89				
							APPLICANT Molnar, C. E.					
CLAIMS												
	AS FILED		AFTER 1st AMENDMENT		AFTER 2nd AMENDMENT							
	IND.	DEP.	IND.	DEP.	IND.	DEP.						
1	1						61					
2		1					62					
3	1						63					
4		1					64					
5	1						65					
6		1					66					
7		1					67					
8		2					68					
9	1						69					
10		1					70					
11		1					71					
12		2					72					
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40							100					
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46												
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48												
49												
50												
TOTAL IND.	4						TOTAL IND.					
TOTAL DEP.	10						TOTAL DEP.					
TOTAL CLAIMS	14						TOTAL CLAIMS					

PTO-1350 (3-78)

\*MAY BE USED FOR ADDITIONAL CLAIMS OR AMENDMENTS

U.S. DEPARTMENT OF COMMERCE  
Patent and Trademark Office

SMS003195



BUMBAUGH, GRAVES, DONOHUE & RAYMOND  
30 ROCKEFELLER PLAZA  
New York, N.Y. 10112

Co. Div. Appl. Trans.

TELEPHONE 212 404-2800  
FACSIMILE 212 404-2800  
CABLE COUNTESS  
TELEX 420000

Our File No. 27515-I 539/15132  
Anticipated Classification  
Class Subclass

IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

The Commissioner of Patents and Trademarks  
Washington, D.C. 20231

Sir:

This is a request for the filing of a

(X) continuation ( ) divisional  
application under 37 CFR 1.60 of pending prior application Serial No. 254,353  
filed on October 6, 1988 of  
Charles E. Melcher  
Inventor(s)

for LUTETIUM ORTHOSILICATE SINGLE CRYSTAL SCINTILLATOR DETECTOR  
Title of Invention

1. ( ) Enclosed is a copy of the prior application as originally filed and an affidavit or declaration verifying it as a true copy.
2. ( ) Small entity status of this application under 37 CFR 1.9 and 1.27 has been established by a verified statement previously submitted.
3. ( ) A verified statement to establish small entity status under 37 CFR 1.9 and 1.27 is enclosed.
4. ( ) The filing fee is calculated below:

FOR	(Col. 1)		(Col. 2)		SMALL ENTITY		OTHER THAN A	
	NO.	FILED	NO.	EXTRA	RATE	FEE	RATE	FEE
Basic Fee	11111111		11111111		11111	\$185	11111	\$370
Total Claims	12	-20 = *	0		x6		x12	= 0
Ind. Claims	4	-3 = *	1		x18		x36	= 36
(2) Multiple Dependent Claims Presented					+60	=	+120	= 120
					TOTAL = \$		TOTAL = \$	526
* If the difference in Col. 1 is less than zero, enter "0" in Col. 2.								

Rev. 4/89

SMS003196

Cr &amp; Div. Appln: Trans.

Our File No. 27515-I 539/15132

5. ☐ An Extension of Time to respond to the rejection dated \_\_\_\_\_ is requested. The required fee, indicated below, is enclosed herewith.

Extension for response (check only one):

	<u>SMALL ENTITY</u>		<u>OTHER THAN A SMALL ENTITY</u>	
Within first month	<input type="checkbox"/>	\$ 31	<input type="checkbox"/>	\$ 62
Within second month	<input type="checkbox"/>	90	<input type="checkbox"/>	180
Within third month	<input type="checkbox"/>	215	<input type="checkbox"/>	430
Within fourth month	<input type="checkbox"/>	340	<input type="checkbox"/>	680

(Check and complete the next item, if applicable)

- 6a. ☐ An extension for \_\_\_\_\_ months has already been secured and the fee paid therefor of \$ \_\_\_\_\_ is deducted from the total fee due for the total months of extension now requested.

Extension fees due with this request \$ \_\_\_\_\_ or

- 6b. ☒ In the event that an extension of time is required, this conditional petition is being made to provide for the possibility that applicant has inadvertently overlooked the need for a petition and fees for extension of time.

- 6c. ☐ TOTAL FEE DUE IS

Filing fees \$ 526.00

Extension fee (if any) \$ \_\_\_\_\_

TOTAL FEE DUE \$ 526.00

7. ☒ The Commissioner is hereby authorized to charge any additional filing fees required under 37 CFR 1.16 and 1.17 associated with this communication or credit any overpayment to Deposit Account No. 02-4377. Two copies of this sheet are enclosed.

8. ☐ Cancel claims \_\_\_\_\_

Co. Div. Appl. Trans.

Our File No. 27515-I 539/15132

- 9a. ☐ Amend the specification by inserting before the first line the sentence:  
This application is a  
☐ continuation ☐ division  
of application Serial No. \_\_\_\_\_, filed on \_\_\_\_\_.
- 9b. ☐ A preliminary amendment is enclosed.
- 10a. ☒ Transfer the drawings from the prior application to this application and abandon said prior application as of the filing date accorded this application.
- 10b. ☐ Formal drawings are enclosed.
- 10c. ☒ Informal drawings are enclosed. (Formal drawings will be forwarded later.)
11. ☒ The prior application is assigned to  
Schlumberger Technology Corporation  
Our File No. 27515 539/11009

12. ☒ a. ☒ The Power of Attorney to our firm or to its members appears in the original papers of the prior application.
- b. ☐ A revocation of the Power of Attorney in the prior application to  
\_\_\_\_\_  
(Name, Reg. No. and Address)  
\_\_\_\_\_  
and a new Power of Attorney are enclosed.
- c. ☐ Since the Power does not appear in the original papers, a copy of the Power in the prior application is enclosed.
- d. ☐ Recognize as associate attorney  
\_\_\_\_\_  
(Name, Reg. No. and Address)  
\_\_\_\_\_

Co. & Div. Appln. Trans.

Our File No. 27515-Y 539/15132

13. ( ) Applicant claims priority in this application under 35 USC 119 on application Serial No. \_\_\_\_\_, filed in \_\_\_\_\_. A certified copy of that application was on \_\_\_\_\_ filed in the parent application Serial No. \_\_\_\_\_ on \_\_\_\_\_.
14. (X) A second duplicate copy of this letter is enclosed for filing in the original application file.
15. (X) Please address all further communications for  
BRUMBAUGH, GRAVES, DONOHUE & RAYMOND  
30 Rockefeller Plaza  
New York, New York 10112

BRUMBAUGH, GRAVES, DONOHUE & RAYMOND

Aug. 4, 1989  
Date

By Arthur S. Tanser  
Arthur S. Tanser  
PTO Registration No. 18,839

"Express Mail" Label No. B86986677  
Date of Deposit August 4, 1989  
I hereby certify that this paper is being deposited with the United States Postal Service "Express Mail Post Office to Addressee" service under 37 CFR 1.10 on the date indicated above and is addressed to the Commissioner of Patents and Trademarks, Washington, D.C. 20231.

Calvin Teegarden  
(Type or print name of person mailing paper or fee)  
Cal Teegarden  
(Signature of person mailing paper or fee)





PATENT  
27515-I 539/15132

07289502

IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

CERTIFICATE OF EXPRESS MAIL UNDER 37 C.F.R. 1.10

Applicant : Charles L. Melcher  
Serial No. : not yet assigned  
Filing Date : (not yet assigned)  
Title of Invention : LUTETIUM Orthosilicate Single  
Crystal Scintillator Detector  
Title of Paper :  
Continuation Application

"Express Mail" Mailing Label  
Number BB6986677

Date of Deposit August 4, 1989

I hereby certify that this paper or fee is being  
deposited with the United States Postal Service "Express  
Mail Post Office to Addressee" service under 37 C.F.R. 1.10  
on the date indicated above and is addressed to the Commis-  
sioner of Patents and Trademarks, Washington, D.C. 20231.

Calvin Teeegården  
(Typed or Printed Name of Person  
mailing paper or fee)

Cal Teeegården  
(Signature of Person mailing  
paper or fee)

SMS003200



UNITED STATES DEPARTMENT OF COMMERCE  
Patent and Trademark Office

Address: COMMISSIONER OF PATENTS AND TRADEMARKS  
Washington, D.C. 20231

SERIAL NUMBER	FILING DATE	FIRST NAMED APPLICANT	ATTY. DOCKET NO.
07/389,502	8-4-89	Charles L. Melcher	27515-1 539/15132

Brunbaugh, Graves, Donchue & Raymond  
30 Rockefeller Plaza  
New York, N.Y. 10112

EXAMINER	
ART UNIT	PAPER NUMBER
	2

DATE MAILED: 8-18-89

IF NO RESPONSE TO THIS NOTICE IS RECEIVED WITHIN FORTY-FIVE DAYS, A  
FORMAL REQUIREMENT WILL BE ISSUED

The subject matter of this application appears to:

☒ be "useful in the production or utilization of special nuclear material or atomic energy" as recited in 42 U.S.C. 2182 (Department of Energy (DOE)).

☐ have significant utility in the conduct of aeronautical and space activities" as recited in 42 U.S.C. 2457 (National Aeronautics and Space Administration (NASA)).

Accordingly, no patent can issue on this application unless applicant(s) file a statement (under oath or in the form of a declaration as provided by 37 CFR 1.60) setting forth (1) the full facts concerning the circumstances under which the invention was made and conceived and (2) the relationship (if any) of the invention to the performance of any work under any contract or other arrangement with the Agency (ies) noted above. On the reverse side of this form is an example of an acceptable format for this statement. The language appearing in paragraphs III and/or IV of the example must appear if applicant is attempting to establish that no relationship (under item 2 above) exists.

If the invention disclosed in this application was developed under a contract, grant or cooperative agreement between the Agency indicated above and a person, small business or non-profit organization and rights to the invention have been determined by specific reference to 35 U.S.C. 202 in the contract, grant or cooperative agreement, then applicant need not submit the statement described above. Instead, applicant may file a verified statement (under oath or in the form of a declaration, 37 CFR 1.60) setting forth the information required by 35 U.S.C. 202(c)(6).

IF NO STATEMENT HAS BEEN RECEIVED WITHIN FORTY-FIVE DAYS OF THE MAIL DATE INDICATED ABOVE, a formal requirement for statement will then be issued. No provision is made for extension of the statutory thirty-day period for response to the formal requirement and the penalty for failure to file an acceptable and timely statement is abandonment of the application. Therefore, applicants are strongly encouraged to submit a statement at this time in order to avoid the issuance of a formal requirement.

IT IS IMPORTANT TO NOTE that the statement must accurately represent the property rights situation of the claimed invention (if and when the application is found allowable. Thus, if during prosecution before the examiner, the claimed invention is so altered or the property rights situation so changed as to impact the accuracy of a statement submitted earlier, a supplemental statement must be filed. Failure to submit such additional information where appropriate may be considered a false representation of material facts and render the patent owner vulnerable to loss of patent rights and other sanctions as set forth in the statutes. The PTO will not review allowed applications for this possibility. The responsibility for complying with the statutes rests with the applicants.

Any questions regarding this requirement should be directed to Licensing and Review at (703) 557-3011

PLEASE DIRECT ALL COMMUNICATIONS RELATING TO THIS MATTER TO THE  
ATTENTION OF LICENSING AND REVIEW

FORM4PTOL-450

U.S. DEPARTMENT OF COMMERCE  
Patent and Trademark Office

SMS003201

The following is an example of an acceptable property rights statement. Statements of this type are, of course, only suitable for situations in which NO Agency funds or other considerations were involved in the making or conception of the invention. While this example is in the form of a declaration, a sworn document is equally acceptable.

I (We) \_\_\_\_\_  
citizens of \_\_\_\_\_  
residing at \_\_\_\_\_  
do declare:

That I (we) made and conceived the invention described and claimed in patent application:

Serial Number \_\_\_\_\_ filed in the United States of America on \_\_\_\_\_  
dated \_\_\_\_\_

(Check and complete either I or II below)

(Check III and/or IV below as appropriate)

☐ I. (For Inventors Employed by an Organization) That I (we) made and conceived this invention while employed by \_\_\_\_\_. That the invention is related to the work I am (we are) employed to perform and was made within the scope of my (our) employment duties; That the invention was made during working hours and with the use of facilities, equipment, materials, funds, information and services of \_\_\_\_\_. Other relevant facts are \_\_\_\_\_.

That to the best of my (our) knowledge and belief (and/or) based upon information provided by \_\_\_\_\_ of \_\_\_\_\_:

—OR—

☐ II. (For Self-Employed Inventors) That I (we) made and conceived this invention on my (our) own time using only my (our) own facilities, equipment, materials, funds, information and services. Other relevant facts are \_\_\_\_\_.

That to the best of my (our) knowledge and belief:

☐ III. The invention was not made or conceived in the course of, or in connection with, or under the terms of any contract, subcontract or arrangement entered into with or for the benefit of the United States Atomic Energy Commission or its successors, Energy Research and Development Administration or the Department of Energy.

—AND/OR—

☐ IV. The invention was not made (conceived or first actually reduced to practice) under nor is there any relationship of the invention to the performance of any work under any contract of the National Aeronautics and Space Administration.

The undersigned inventor(s) declare further that all statements made herein of his or her (their) own knowledge are true and that all statements made on information and belief are believed to be true and further that these statements are made with the knowledge that willful false statements and the like so made are punishable by fine or imprisonment, or both, under Section 1001 of Title 18 of the United States Code and that such willful false statements may jeopardize the validity of the application or any patent issuing thereon.

Inventor's Signature: \_\_\_\_\_

Post Office Address: \_\_\_\_\_

Date: \_\_\_\_\_

Inventor's Signature: \_\_\_\_\_

Post Office Address: \_\_\_\_\_

Date: \_\_\_\_\_

SMS003202

27515-I 539/15132  
PATENT

IN THE UNITED STATES PATENT AND TRADEMARK OFFICE



RECEIVED  
SEP 11 1989  
LAW OFFICE 7 REVIEW

CERTIFICATE OF MAILING UNDER 37 C.F.R. 1.8(a)

Declaration Under Section 152  
of the Atomic Energy Act of Serial No. 07/389,502  
Paper ~~1954-42 U.S.C. Section~~  
2182 Filing Date Aug. 4, 1989

I hereby certify that this paper is being deposited  
with the United States Postal Service as first class mail in  
an envelope addressed to:

Commissioner of Patents and Trademarks  
Washington, D.C. 20231

on September 5, 1989  
Date of Deposit

September 5, 1989  
Date of Signature

Arthur S. Tanger  
Attorney Signature

Arthur S. Tanger  
Attorney Name

18,839  
Registration No.

SMS003203

27515-I-539/15132  
PATENT

#3  
(152)

IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

Applicant : Charles L. Melcher  
Serial No. : 07/389,502  
Filed : August 4, 1989  
For : LUTETIUM ORTHOSILICATE SINGLE  
CRYSTAL SCINTILLATOR DETECTOR

RECEIVED  
SEP 11 1989  
LUTETIUM A. PREVIEW

September 5, 1989

Hon. Commissioner of Patents  
and Trademarks  
Washington, D.C. 20231

Sir:

Responsive to the Notice dated August 18, 1989,  
transmitted herewith is an executed Declaration under Section 152  
of the Atomic Energy Act of 1954 42 U.S.C. 2182.

Respectfully submitted,

  
Arthur S. Tenser  
PTO Registration No. 18,039

Attorney for Applicant  
(212) 408-2542

Enclosure

SMS003204

27515-I-539/15132

PATENT

IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

RECEIVED  
SEP 11 1989  
LICENSING & REVIEW

Applicant : Charles L. Melcher  
Serial No. : 07/389,502  
Filed : August 4, 1989  
For : LUTETIUM ORTHOSILICATE SINGLE  
CRYSTAL SCINTILLATOR DETECTOR

DECLARATION UNDER SECTION 152  
OF THE ATOMIC ENERGY ACT OF 1954  
(42 U.S.C. §2182)

Hon. Commissioner of Patents  
and Trademarks  
Washington, D. C. 20231

ATTENTION: Licensing and Review Section

I, CHARLES L. MELCHER, state that I am the inventor  
in the matter of the above-identified application and I  
declare that:

At the time I made the invention which is the sub-  
ject matter of the above-identified application, I was an  
employee of the assignee of the above-identified application,  
Schlumberger Technology Corporation. As an employee of  
Schlumberger Technology Corporation, my job as a member of the  
professional staff included designing and developing tech-  
niques and apparatus for investigating the characteristics of  
earth formations surrounding an oil or gas well.

The invention to which the above-identified appli-  
cation is addressed was made by me during the course of my  
assigned work for Schlumberger Technology Corporation, the  
assignee of the above-identified application. A description  
of this invention is recorded in various records and drawings  
of Schlumberger Technology Corporation, with these records and

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SMS003205

27515-I-539/15132

drawings serving as the basis for the disclosure in the above-identified application. The invention to which the above-identified application is addressed was made by me with funds, facilities, and equipment provided by Schlumberger Technology Corporation. Moreover, the invention was made during my regular working hours and in the course of my regular employment with Schlumberger Technology Corporation.

To the best of my knowledge and belief, the invention was not made or conceived in the course of, or in connection with, or under the terms of any contract, subcontract, or arrangement entered into with or for the benefit of the United States Atomic Energy Commission or its successors: the Energy Research and Development Administration or the Department of Energy.

The undersigned declares that all statements made herein of his own knowledge are true and that all statements made on information and belief are believed to be true; and further that these statements were made with the knowledge that willful false statements and the like so made are punishable by fine or imprisonment, or both, under Section 1001 of Title 18 of the United States Code and may jeopardize the validity of the application or any patent issuing thereon.

8-29-89  
Date

Charles L. Melcher  
CHARLES L. MELCHER  
49 Lampost Road  
West Redding, CT 06896



UNITED STATES DEPARTMENT OF COMMERCE  
Patent and Trademark Office  
Address: COMMISSIONER OF PATENTS AND TRADEMARKS  
Washington, D.C. 20231

SERIAL NUMBER	FILING DATE	FIRST NAMED APPLICANT	ATTORNEY DOCKET NO.
07/389,502		CHARLES L. MELCHER	
<input type="checkbox"/> Plus the subject matter described in the material accompanying the petition. (Applies only if box is marked)			
LUTETIUM ORTHOSILICATE SINGLE CRYSTAL SCHMILLATOR DETECTOR			
EXAMINER			
ART UNIT		PAPER NUMBER	

DATE MAILED: Oct. 31/97

ARTHUR S. TENSER

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DATE OCT 31 1997

Commissioner of Patents and Trademarks

*Am. 8*  
This license empowers the filing, the examination and the authorization of the filing of a foreign application or applications on the subject matter identified above, subsequent forwarding of all dispatches and formal papers and the prosecution of such application or applications.  
This license does not empower the filing of any applications, amendments, supplements or continuations originating in this country which disclose inventions, modifications, or variations not disclosed in the subject matter identified above.  
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The grant of this license does not in any way lessen the responsibility of the licensee for the security of the subject matter as imposed by any Government control or the provisions of existing laws relating to espionage and the national security or the export of technical data. Licensees should apprise themselves of current regulations, especially with respect to certain countries, of other agencies, particularly the Office of Munitions Control, Department of State (with respect to Arms, Munitions and Implements of War); the Bureau of Trade Regulation, Office of Export Administration, Department of Commerce; and the Department of Energy.

LICENSE FOR FOREIGN FILING  
(Title 35, United States Code (1962) Sections 184, 185, 186)

PTO 200 (Rev. 5-81)

SMS003207



2 5-1 539/15132  
PATENT

IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

Applicant: Charles L. Melcher

Serial No.: 07/389,502

Examiner: Cooper Not Assigned

Filed : August 4, 1989

Group Art Unit: 356-CP3

For : LUTETIUM ORTHOSILICATE SINGLE  
CRYSTAL SCINTILLATOR DETECTOR

February 16, 1990

PRELIMINARY AMENDMENT

Hon. Commissioner of Patents and Trademarks  
Washington, D.C. 20231

Sir:

Preliminary to examination, please amend the  
above-identified application as follows:

In the Specification

Page 3, line 25, after the "." insert - U.S.

Application Serial No. 149,953 issued as U.S. Patent No.  
4,883,956, granted November 28, 1989.--.

R E M A R K S

By the foregoing amendment, the status of the co-  
pending application of the assignee is brought up to date.

As a matter of convenience we enclose a copy of the  
amendment and attached documents filed in applicant's parent  
application Serial No. 07/254,353, now abandoned. Also  
attached is a copy of the Examiner's Interview Summary Record  
dated May 2, 1989 and the Notice of Allowability with  
attached Examiner's Amendment dated May 9, 1989. It is

SMS003208

2,565-1 539/15132  
PATENT

respectfully requested that these documents be included in  
the file of the present application.

Respectfully submitted,



Arthur S. Tenner  
Reg. No. 18,839

Attorney for Applicant  
(212) 400-2542

Enclosures

SMS003209

27515-539/11889

PATENT

98 FEB 20 6:11 PM  
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 IN THE UNITED STATES PATENT AND TRADEMARK OFFICE  
 MAIL 110

Applicant : Charles L. Melcher  
 Serial No. : 07/254,353 Examiner : J. Cooper  
 Filed : October 6, 1988 Group Art Unit: 113  
 For : LUTETIUM ORTHOSILICATE SINGLE  
 CRYSTAL SCINTILLATOR

I hereby certify that this paper is being  
 deposited with the U. S. Postal Service as  
 First Class Mail in an envelope addressed to:  
 Commissioner of Patents and Trademarks,  
 Washington, DC 20231, on February 24, 1989

Richard G. Berkley	25,455
Attorney Name	Registration No.
<i>Richard G. Berkley</i>	February 24, 1989
Signature	Date of Signature

AMENDMENT

Hon. Comm. of Patents and Trademarks  
 Washington, D. C. 20231

Sir:

In response to the Office Action dated November 28,  
 1988 in the above-identified application, please amend the  
 application as follows:

In the Claims:

Please amend claims 1-4 as follows:

--1. (Amended) A scintillator for use in a gamma ray  
 or x-ray (like radiation) detector, comprising a transparent  
 single crystal of cerium-activated lutetium oxyorthosilicate  
 having the general formulation  $\text{Ce}_{2x}\text{Lu}_{2(1-x)}\text{SiO}_5$ , where x is  
 [the concentration in the melt from which the crystal is  
 pulled and is] within the range of from approximately {0.001}  
 $2 \times 10^{-4}$  to approximately {0.1}  $3 \times 10^{-2}$ .---

SMS003210

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--2. (Amended) The scintillator of claim 1 wherein x is within the range of approximately  $[0.005] \underline{1 \times 10^{-3}}$  to approximately  $[0.015] \underline{4.5 \times 10^{-3}}$ ;

--3. (Amended) A gamma ray or x-ray [like radiation] detector, comprising:

a scintillator composed of a transparent single crystal of cerium-activated lutetium oxyorthosilicate having the general formulation  $\text{Ce}_{2x}\text{Lu}_{2(1-x)}\text{SiO}_5$ , where x is [the concentration in the melt from which the crystal is pulled and is] within the range of from approximately  $[0.001] \underline{2 \times 10^{-4}}$  to approximately  $[0.1] \underline{3 \times 10^{-2}}$ , and

a photodetector optically coupled to the scintillator for producing an electrical signal in response to the emission of a light pulse by the scintillator.---

--4. (Amended) The detector of claim 3 wherein x is within the range of approximately  $[0.005] \underline{1 \times 10^{-3}}$  to approximately  $[0.015] \underline{4.5 \times 10^{-3}}$ ---

#### R E M A R K S

Claims 1-4 have been rejected for indefiniteness and for obviousness over the prior art. Reconsideration and withdrawal of these rejections are respectfully requested.

The indefiniteness rejection is based on alleged indefiniteness in (1) the use of the term "or like" to refer to radiation other than gamma ray radiation to which the scintillator responds, (2) the definition of the cerium content of the crystal in terms of the cerium concentration in the melt from which the crystal is pulled, and (3) the use of the lan-

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guage "in the melt from which the crystal is pulled". Claims 1-4 have been amended to eliminate all three grounds of rejection. The first ground has been eliminated by deleting the term "like radiation" from independent claims 1 and 3 and substituting --x-ray-- in place thereof. Clear support for the recitation of x-ray detection is found at page 5, line 29 - page 6, line 2. No issue of new matter arises. Also, no indefiniteness results from defining the scintillator as responsive to "gamma ray or x-ray" radiation, inasmuch as gamma rays and x-rays differ only as to origin and are essentially indistinguishable insofar as the scintillator is concerned.

The second and third grounds for the indefiniteness rejection has been eliminated by amending claims 1-4 to delete the reference to the melt and the melt concentrations and to define the cerium concentration by reference to the crystal alone. At page 11, lines 23-24, of the specification, it is disclosed that the cerium concentration in the crystal is on the order of 20%-30% of that in the melt. That is to say, approximately 20%, on the low side, and 30%, on the high side, of the cerium in the melt will be substituted for the lutetium in the LSO crystal. Applying 20% to the lower limit of x in the formula of claims 1 and 3 and 30% to the upper limit of x in that formula yields a range of x of from approximately  $2 \times 10^{-4}$  to approximately  $3 \times 10^{-2}$  as the range of cerium concentration in the crystal. Claims 1 and 3 have been amended to recite this range. Claims 2 and 4 have similarly been amended to recite the preferred range of x in the crystal. In light of the clear support in the specification from these amendments, no new matter is introduced, and grounds (2) and (3) for the indefiniteness rejection are removed.

27515-539/11889

The obviousness rejection is based on the disclosure of the Matsushita patent document in combination with the disclosures of the Takagi et al., patent and the Watanabe et al. patent. The Examiner acknowledges that while Matsushita discloses a cerium-activated lutetium oxyorthosilicate (LSO) luminescent material, he fails to disclose such material in single crystal form or its use as a gamma ray/x-ray detector. The Examiner concludes, however, that it would have been suggested (and thus obvious) to one of ordinary skill in the art from Takagi et al., which discloses a cerium-activated gadolinium oxyorthosilicate (GSO) single crystal gamma ray detector, and Watanabe, which allegedly discloses the "common use of gadolinium and lutetium" in cerium-activated rare earth oxyorthosilicate luminescent material, to use the luminescent material of Matsushita in single crystal form in a radiation detector. We respectfully submit that the references relied on, whether taken singly or in combination, fail to disclose or suggest either the claimed scintillator or its use as a gamma ray/x-ray detector, and that the rejection should therefore be withdrawn.

In addition to its failure to disclose either the single crystal form of LSO or the use of LSO as a gamma ray/x-ray detector, Matsushita is deficient in at least two other important respects as well: it includes no disclosure at all, one, that cerium-activated LSO in any form is responsive to gamma ray/x-ray excitation, and, two, that cerium-activated LSO could even be grown in single crystal form or that, if it were, that it would have the transparent properties or scintillation properties required for a gamma ray/x-ray detector. In fact, all that Matsushita does disclose is a particular

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process for producing pure white electron beam (EB) excitable and ultraviolet (UV) excitable cerium-activated powder phosphors for use in flying-spot tubes and plasma displays. (A translation of Matsushita is attached as Exhibit A.) There is no mention whatsoever of the single crystal form, transparency, or response (useful or otherwise) to gamma ray/x-ray excitation. Certainly, therefore, there is no suggestion in Matsushita that the claimed invention should or could be made.

Although Takagi et al. do disclose the use of a cerium-activated GSO single crystal scintillator as a gamma ray detector, they do not disclose that any other rare earth oxyorthosilicate material would be suitable for use as a cerium-activated single crystal gamma ray or x-ray detector. Indeed, they criticize the use of the rare earth oxyorthosilicate  $Y_2SiO_5$  (yttrium oxyorthosilicate) for that purpose. (See Col. 2, line 10 et seq. of the Takagi et al. patent)

Beyond the criticism of yttrium, there are other good reasons why those skilled in the art would not have been led by Takagi et al. to conclude that the white phosphor form of L60 disclosed by Matsushita should, or even could, be useful in the single-crystal form as a gamma ray/x-ray detector. First, although the rare earth silicates are similar chemically, their optical and luminescent properties are known to differ widely and unpredictably. For example, ytterbium and lutetium are adjacent to one another in the periodic chart and are quite similar chemically. Yet their optical properties are dramatically different even at the phosphor stage, let alone the crystal stage. Second, the optical and luminescent properties of the single-crystal form are not predictable from the phosphor form. Third, the conversion efficiency of inci-

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dent photons to light output in a luminescent material depends upon the energy of the incident radiation and the loss mechanism in the material. Hence that a material luminesces in response to EB or UV excitation does not indicate how the material would respond to gamma ray or x-ray excitation. Fourth, the ionic size of cerium is about twice that of lutetium, while cerium and gadolinium are of approximately the same ionic size. Thus the fact that cerium ions substitute for gadolinium ions in the growth of a GSO single crystal does not predict that cerium ions would be taken up in sufficient concentration or with the proper distribution in an LSO single crystal to provide a useful gamma ray/x-ray scintillator.

Furthermore, the prosecution record of the Takagi et al. patent provides convincing objective evidence that the disclosure of a cerium-activated GSO single crystal scintillator does not suggest the utility of a cerium-activated LSO single crystal scintillator. In the parent application (Serial No. 462,227) and again in the continuation application (Serial No. 787,076), Takagi submitted a Declaration describing experiments that he had conducted with two known scintillators under gamma ray excitation,  $Y_3Al_5O_{12}:Ce$  and  $YAlO_3:Ce$ , and two like compounds except that the yttrium (Y) was replaced by gadolinium (Gd). (A copy of the Declaration from Application Ser. No. 787,076 is attached as Exhibit B.) In the latter two cases, the gadolinium compounds did not exhibit scintillation under gamma ray (γ) excitation. Based on these results, Takagi concluded that "even when a compound containing Y does serve as a scintillator for γ ray irradiation, it cannot be estimated whether or not another compound in which Y is substituted with Gd can also serve as a scintillator for



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x-ray irradiation". (See Exhibit A, p. 10, emphasis added) Thus, Takagi himself is of record as stating that the scintillation properties of one rare earth compound do not afford a basis for predicting the scintillation properties of another rare earth compound.

It cannot be said, therefore, that one skilled in the art would be led by Takagi et al. to conclude that the EB and UV-excited LSO white phosphor of Matsushita should (or even could) be grown into a transparent, single-crystal form or that, if it were, that it would possess the luminescent properties required for a gamma ray/x-ray detector. Rather, the Takagi et al. disclosure and prosecution record, taken as a whole, actually lead away from that suggestion.

As previously noted, the Watanabe et al. patent has been cited by the Examiner as teaching the common use of gadolinium and lutetium in a cerium activated rare earth oxyorthosilicate luminescent material. What Watanabe et al. in fact disclose is a rare earth (Y, Ln, Gd, or Lu) oxyorthosilicate which is co-activated by cerium and terbium to produce visible green rays when excited by ultraviolet (UV) radiation whose wavelengths lie within a broad range. The high efficiency of the phosphor is attributed to the transmission to the activator Tb<sup>3+</sup> of the energy of UV rays absorbed in the activator Ce<sup>3+</sup>. (See Col. 2, lines 55-66) The purpose is to provide a green-light emitting phosphor for use in fluorescent lamps.

At the most, therefore, Watanabe et al. disclose the common use of GSO and LSO as Ce and Tb - coactivated phosphors for green-light emission under broad band UV excitation. As this involves an altogether different physical structure and

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an altogether different excitation and scintillation mechanism than those of the claimed invention, it neither discloses nor suggests anything concerning the interchangeability of GSO and LSO as Co-activated single crystal scintillators for gamma/x-ray excitation. Hence, Watanabe et al. would not lead one skilled in the art to modify the Matsushita phosphor in the manner proposed in the rejection.

The art completely lacks any disclosure of a transparent, single crystal cerium-activated lutetium oxyorthosilicate scintillator. Furthermore, for the reasons given above, the art of record lacks any suggestion of either the claimed scintillator or its use as a gamma ray/x-ray detector. As recently stated by the Court of Appeals for the Federal Circuit, obviousness "cannot be established by combining the teachings of the prior art to produce the claimed invention, absent some teaching or suggestion supporting the combination." In re Fine, 5 U.S.P.Q. 2d 1596, 1599. And "the teachings of references can be combined only if there is some suggestion or incentive to do so (Id. at 1599, emphasis in original). Here, as in In re Fine, the prior art contains no such teaching, suggestion or incentive. We submit, therefore, that claims 1-4, as amended, define patentably over the prior art and are allowable.

Pursuant to 37 C.F.R. §§1.97 and 1.98, applicant encloses herewith a copy of each of the references listed on the attached form PTO-1449 and requests that it be considered and made of record in the application. The references were not earlier cited because this application was examined and acted upon by the Examiner within two months of its filing date, which was well before either of the two recommended

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deadlines for filing an Information Disclosure Statement provided in 37 C.F.R. §1.97.

Buisson et al. disclose the crystallographic parameters of lutetium oxyorthosilicate powder along with those for other rare earth oxyorthosilicates.

Gomes de Mesquita et al. disclose cerium-activated lutetium oxyorthosilicate powders for use as cathode-ray phosphors.

Anan'eva et al. describe the growth by the Czochralski method of undoped single crystals of oxyorthosilicates of lanthanides from gadolinium to lutetium, and of yttrium oxyorthosilicate.

Holaa et al. describe the optical properties of  $\text{Eu}^{3+}$ -activated rare earth oxyorthosilicate powders, wherein the rare earth elements were gadolinium, yttrium and lutetium.

Takagi et al. relates to the same general subject matter of the Takagi et al. patent already of record, i.e., a cerium-activated gadolinium oxyorthosilicate single crystal for use in position computed tomography.

Brandle et al. describe the growth of crystals of the type  $\text{Ln}_2\text{SiO}_5$  (Ln is a lanthanide) by the Czochralski method. Of the four original lanthanides selected for evaluation ( $\text{Lu}_2\text{SiO}_5$ ,  $\text{Ce}_2\text{SiO}_5$ ,  $\text{Gd}_2\text{SiO}_5$ , and  $\text{Y}_2\text{SiO}_5$ ) only two ( $\text{Gd}_2\text{SiO}_5$  and  $\text{Y}_2\text{SiO}_5$ ) were found suitable for Czochralski growth experiments.

Rabinovich et al. disclose the preparation of sols and gels of  $\text{Tb}^{3+}$ -doped yttrium orthosilicate for use as cathodoluminescent films.

Blasse et al. describe the luminescence properties of a large number of phosphors or phosphor families. The

27515-539/11889

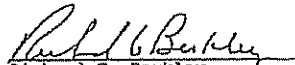
authors conclude that while much is known about phosphors, much is not well known and is subject to empirical prediction, if at all.

Ishii et al. (English language) and the last listed Japanese-language paper (of which a translation is not available) both appear to relate to the cerium-activated GSO crystal scintillator of the Takagi patent of record. They are being submitted for completeness.

None of these additional references, whether taken alone or in combination, disclose or suggest the cerium-doped LSO single crystal scintillator of the present invention nor its use as a gamma ray/x-ray detector. Accordingly, the claims are patentably distinct from these references as well.

The application is believed to be in readiness for allowance in all respects. A favorable action is respectfully requested.

Respectfully submitted,

  
Richard G. Berkley  
Reg. No. 25,465  
Attorney for Applicant  
(212) 400-2554

Encl.

PTO-102X  
Rev. 11-82

## FILING RECEIPT

UNITED STATES DEPARTMENT OF COMMERCE  
Patent and Trademark Office  
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Washington, D.C. 20231

SERIAL NUMBER	FILING DATE	EXP. APT. UNIT	FL. FEE REC'D	ATTORNEY DOCKET NO.	DRWS	TOT. CL.	IND. CL.
07/254,353	10/06/88	256	\$ 340.00	27515-539/11	1	4	2

BRUMBAUGH, GRAVES, DONOHUE & RAYMOND  
30 ROCKEFELLER PLAZA  
NEW YORK, NY 10112

Receipt is acknowledged of the patent application identified herein. It will be considered in its order and you will be notified as to the examination thereof. Be sure to give the U.S. SERIAL NUMBER, DATE OF FILING, NAME OF APPLICANT, and TITLE OF INVENTION when inquiring about this application. Fees transmitted by check or draft are subject to collection. Please verify the accuracy of the data presented on this transmittal.

Applicant(s) CHARLES L. MELCHER, WEST REDDING, CT.

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TITLE  
LUTETIUM ORTHOSILICATE SINGLE CRYSTAL SCINTILLATOR

PRELIMINARY CLASS: 250

EXHIBIT A

RECEIVED  
BRUMBAUGH, GRAVES,  
DONOHUE & RAYMOND  
1988 NOV 15 PM 1:31

(see reverse)

SMS003220



**Lindner Translations, Inc.**

75 BROADWAY • NEW YORK, N.Y. 10006 • TEL. (212) 759-4510

(19) Patent Office of Japan

**Gazette of Unexamined Patent Applications**

(11) Japanese Laid-Open Patent Application No.: 51-59,079 (1976)  
(43) Date laid open: May 22, 1976  
(21) Patent application No.: 49-134,122 (1974)  
(22) Application date: November 20, 1974  
Request for examination: not filed (Total of 3 pages)

Patent Office Reference No.: 7433 4A

(52) Japanese Cl.: 13(9)C114.2

(51) Int. Cl.<sup>2</sup>: C09K 11/4611  
B01J 31/10

**PATENT APPLICATION (A5)**

November 20, 1974

To: Director of the Patent Office

**1. Title of the Invention:**

Cerium-Activated Lutetium Silicate Phosphor Production Process

**2. Inventor:**

Address: Matsushita Electric Industrial Co., Ltd.  
1006 Kadoma, Kadoma-shi, Osaka Prefecture  
Name: Fumio FUKUSHIMA  
(and 2 others)

### **Lindner Translations, Inc.**

23 RADADHAY • NEW YORK, NY 10006 • TEL. (212) 763-4160

#### **3. Applicant:**

Address: 1006 Kadoma, Kadoma-shi, Osaka Prefecture  
Name: (582) Matsushita Electric Industrial Co., Ltd.  
President: Masaharu MATSUSHITA

#### **4. Agent:**

Address: Matsushita Electric Industry Co., Ltd.  
1006 Kadoma, Kadoma-shi, Osaka Prefecture 571  
Name: (5971) Toshio NAKAO, a registered patent attorney  
(and 1 other)  
Telephone: (03)453-3111 (Patent Division)

#### **5. List of Attached Documents:**

- |                              |          |
|------------------------------|----------|
| (1) Specification            | (1 copy) |
| (2) Diagram                  | (1 copy) |
| (3) Power of Attorney        | (1 copy) |
| (4) Duplicate of Application | (1 copy) |

#### **6. Inventors and Agents Other Than Those Cited Above:**

##### **(1) Inventors:**

Address: Matsushita Electric Industry Co., Ltd.  
1006 Kadoma, Kadoma-shi, Osaka Prefecture  
Name: Yoji FUKUDA

Address: same as above  
Name: Masakazu FUKAI

##### **(2) Agent:**

Address: Matsushita Electric Industry Co., Ltd.  
1006 Kadoma, Kadoma-shi, Osaka Prefecture  
Name: (5152) Shigetaka KURINO, a registered patent attorney

**Lindner Translations, Inc.**

27 BROADWAY • NEW YORK, N.Y. 10006 • TEL. (212) 269-4500

**SPECIFICATION**

**1. Title of the Invention**

Cerium-Activated Lutetium Silicate Phosphor Production Process

**2. Claim**

A process for producing cerium-activated lutetium silicate phosphors having the general formula  $(Lu, Ce)_2SiO_5$ , which process is characterized by final firing in a vacuum or in a reduced-pressure atmosphere.

**3. Detailed Description of the Invention**

The present invention relates to a process for producing cathode ray-excitable or ultraviolet ray-excitable cerium-activated lutetium silicate phosphors.

Cerium-activated lutetium silicate phosphors show promise as high-efficiency flying-spot tube phosphors and also, more recently, as blue phosphors for plasma displays. The peak wavelength in the emission spectrum of these phosphors is near 400 nm, and the luminance is not high. Yet, phosphors for color flying-spot tubes and plasma displays must have a higher luminance.

The aim of the present invention, therefore, is to obtain cerium-activated lutetium silicate phosphors that have a strong emission intensity and a pure white color.



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Up until now, cerium-activated lutetium silicate phosphors have been produced by thoroughly mixing a lutetium compound such as lutetium oxide with a silicate compound such as silicon dioxide, a cerium compound such as cerium oxide, and a fluoride flux such as lutetium fluoride, then firing at a high temperature of about 1000° or more. This firing was generally carried out in air or in a weakly reducing atmosphere. When the cerium concentration in the  $(Lu, Ce)_2SiO_5$  phosphors obtained in this way is 5% or less, the emission peak is near 400 nm and the luminance is low. If the cerium concentration is raised to 5-10%, the long wavelength side of the emission spectrum increases relative to the short wavelength side, but the emission intensity declines markedly and the phosphors take on somewhat of a other color. Curve a in the graph shows the emission spectrum due to electron beam-excitation when the cerium concentration is 3%.

In the process for fabricating cerium-activated lutetium silicate phosphors according to the present invention, the fired product obtained in a normal synthesis process is subjected in final firing in a vacuum or a reduced-pressure atmosphere. Hitherto, phosphors having oxygen acid-base compounds such as  $Lu_2SiO_5$  as the matrix have been fired in either oxidizing atmospheres such as air or oxygen, or in reducing atmospheres such as hydrogen; there are no instance where an attempt has been made to improve the properties (of the phosphor) by treatment in a vacuum or in a reduced-pressure atmosphere. The  $(Lu, Ce)_2SiO_5$

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phosphors obtained through the production process of the present invention, however, have been found to have a higher luminance than prior-art product and also to have an increased emission intensity and a whiter color.

More specifically, the oxides of the component elements are mixed thoroughly with a flux such as lutetium fluoride (which need not necessarily be used), then [the mixture] is fired for several hours in air at a temperature of 1200-1500° C. The fired product thus obtained is ground well, mixed, placed in an evacuated sealed vessel or a reduced-pressure vessel, and fired for several hours at an elevated temperature of at least 900° C. Compared with samples that were not vacuum treated, the phosphors obtained in this way showed an improvement of up to about 50% in the luminance of electron beam-excited emission. In addition, the emission output by a photomultiplier tube 1P21 (S4 photoelectric surface) was improved by about 10%. Plot b in the graph shows the emission spectrum resulting from the electron-beam excitation of cerium-activated  $\text{Lu}_2\text{SiO}_5$  phosphors fired in a vacuum. As is apparent from this graph, the luminance of the phosphors obtained through the present invention can be improved over that of prior-art phosphors. Also, this improvement clearly originates primarily in the increase in emission on the long wavelength side of the peak. The fact that the phosphor color becomes pure white when fired in a vacuum is also a major feature.

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This effect is large when the cerium concentration is high (at least about 5%). An even larger effect of firing in a vacuum is the improvement in the ultraviolet ray-excited emission. It was possible to easily effect an improvement of 1.5-2 fold in the luminance and 1P21 photomultiplier tube output for emission induced by low-pressure mercury lamp (2537 Å) excitation. In high-pressure mercury lamp (3664 Å) excitation, the luminance was about 1.5 times that prior to firing in a vacuum. The following table shows one example of the results of treatment in a vacuum upon the luminance and emission output (1P21 output) for  $\text{Lu}_2\text{SiO}_5$  phosphors when the cerium concentration is 1% and 5%.

Ce concentration	Vacuum firing conditions (temperature, time)	Electron-beam excitation		UV-excitation (2537 Å)	
		1P21 output	Luminance	1P21 output	Luminance
1%	Prior art: N/A	100	100	100	100
	Invention: 1250° C, 4 hrs	110	139	206	199
5%	Prior art: N/A	100	100	100	100
	Invention: 1250° C, 4 hrs	112	151	160	209

As is clear from the above table, compared with prior-art phosphors, the phosphors of the present invention exhibit improved emission output and luminance in both the case of electron-beam excitation and ultraviolet-ray excitation.

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In the case of electron beam-excited emission, the advantageous effects of firing in a vacuum are observed at a firing temperature of about 900° and above. Ultraviolet ray-excited emission is extremely sensitive to the vacuum-firing temperature; even when firing was conducted at 500° C for 30 hours, improvements in the emission output and luminance of about 10% were observed.

Examples of the invention are given below.

Example 1

lutetium oxide ( $\text{Lu}_2\text{O}_3$ )	393.93 g
silicon dioxide ( $\text{SiO}_2$ )	72.11 g
cerium oxide ( $\text{CeO}_2$ )	3.44 g

These were thoroughly mixed in a mortar, placed in a platinum crucible, and heated and fired in air. Firing was carried out at 1300° C for 3 hours. The fired product thus obtained was well ground, following which it was wrapped in platinum sheet and vacuum encapsulated within a quartz ampule. This was fired at 1250° C for 4 hours. The emission output and luminance resulting from the electron-beam excitation of phosphors obtained in this way were respectively 110% and 139% the values obtained for cerium-activated  $\text{Y}_2\text{SiO}_5$  that was not heated in a vacuum. The emission output and luminance resulting from ultraviolet-ray excitation with a low-pressure mercury lamp (2537 Å) were respectively 206% and 103%.

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Example 2

lutetium oxide ( $\text{Lu}_2\text{O}_3$ )	378.01 g
silicon dioxide ( $\text{SiO}_2$ )	72.11 g
cerium oxide ( $\text{CeO}_2$ )	17.21 g

These were thoroughly mixed in a mortar, placed in a platinum crucible, and heated and fired in air. Firing was carried out at  $1450^\circ\text{C}$  for 3 hours. The fired product thus obtained was well ground, following which it was wrapped in platinum sheet, placed in a quartz ampule sealed "on one side," and fired at  $1200^\circ\text{C}$  for 3 hours while evacuating with an exhaust pump. Compared with phosphor that was not fired in a reduced-pressure atmosphere, the phosphor obtained through this procedure showed vast improvements in emission upon electron-beam excitation and ultraviolet-ray excitation.

As is apparent from the above, in the production of cerium-activated lutetium silicate ( $\text{Y, Ce}_2\text{SiO}_5$ ) phosphor, the emission output and luminance can be improved by conducting the final firing in a vacuum or in a reduced-pressure atmosphere,

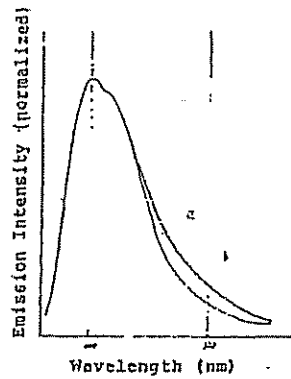
4. Brief Description of the Diagram

The diagram shows the emission spectrum obtained by the electron-beam excitation of cerium-activated lutetium silicate phosphor. Curve a represents phosphor produced by a prior-art method in which vacuum firing is not carried out, while curve b represents phosphor produced by the method of the present invention.

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Name of Agent: Toshio NAKAO, a registered patent attorney  
(and 1 other)





IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

In re the application of

Applicants: K. TAKAGI ET AL

Serial No.: 787,076

Filed: October 15, 1985

Title: GAMMA RAY DETECTOR

Art Unit: 256

Examiner: R. Hanig

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U.S. PATENT & TRADEMARK OFFICE

DECLARATION

I, Kazumasa TAKAGI, residing at 2196-376,  
Hirai Minode Nishitama-gun, Tokyo, Japan, declare and  
say as follows:

1. I am a graduate of the Department of  
metallurgy, Faculty of Engineering, Osaka University,  
Japan and in November 1982, and I received a degree of  
Doctor of Engineering.

2. Since 1971, I have been employed by  
Central Research Laboratory, Hitachi Ltd. and during  
this period of time I have been engaged in research in  
Crystal Growth.

3. During my employment with Central Research  
Laboratory, Hitachi Ltd. I have written the following  
technical articles:

- 1 -

SMS003230

"Improvement in the Scintillation Conversion Efficiency of  $\text{Bi}_4\text{Ge}_3\text{O}_{12}$  Single Crystals" by Kazumasa TAKAGI, Tokumichi FUKAZAWA, ET. AL., in J. Crystal Growth 52, 584-588 (1981), "Cerium-activated  $\text{Gd}_2\text{SiO}_5$  Single Crystal Scintillator" by Kazumasa TAKAGI, Tokumichi FUKAZAWA in Appln. Phys. Lett 42, 43-45 (1983).

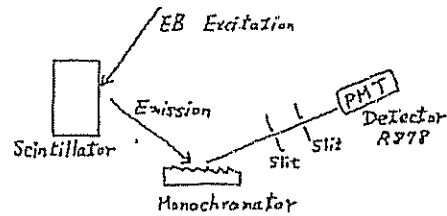
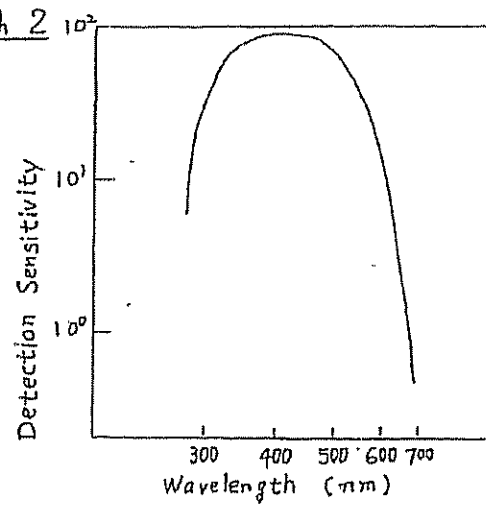
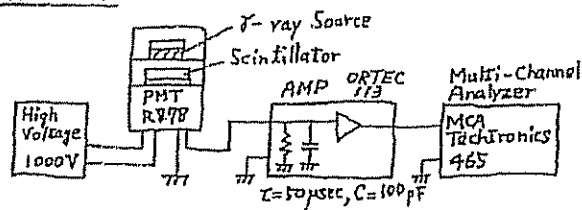
4. I invented, with my colleague Fukazawa, a gamma ( $\gamma$ ) ray detector comprising a scintillator formed of a single crystal of  $\text{Gd}_{2(1-y)}\text{Ce}_y\text{SiO}_5$  (where  $1 \times 10^{-3} \leq y \leq 0.1$ ) and a photodetector for detecting light from said scintillator, and conducted measurements on a scintillator formed of a single crystal of  $\text{Gd}_{1.99}\text{Ce}_{0.01}\text{SiO}_5$ .

#### Experimental:

Single crystals of  $\text{Gd}_{1.99}\text{Ce}_{0.01}\text{SiO}_5$  were grown, shaped and polished to optical grade to form scintillator crystals.

Emission peak wavelength was determined from the emission spectrum measured by a measuring system as shown in Sketch 1. An electron beam (EB) was irradiated on the scintillator crystal of  $\text{Gd}_{1.99}\text{Ce}_{0.01}\text{SiO}_5$  to excite scintillation. Lights emitted from the scintillator crystal were directed toward a grating monochromator and dispersed thereat. The dispersed lights were passed through double slits to select monochromatic lights.



Sketch 1Sketch 2Sketch 3

- 3 -

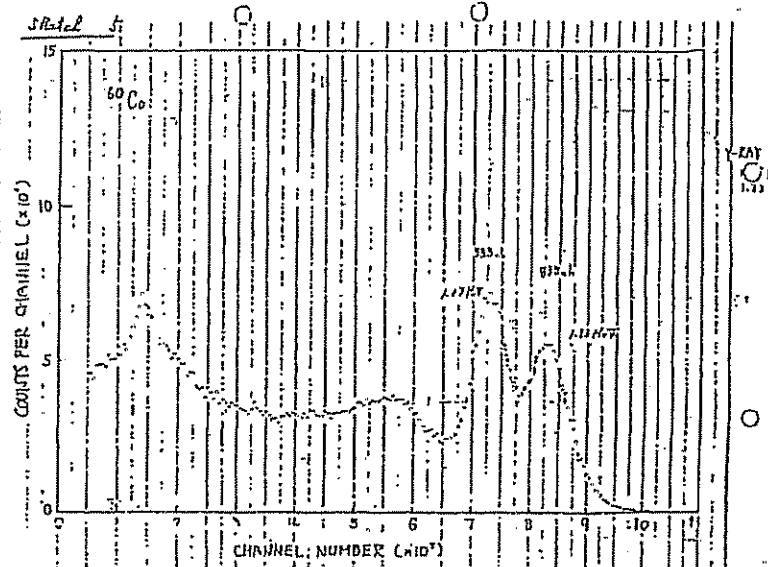
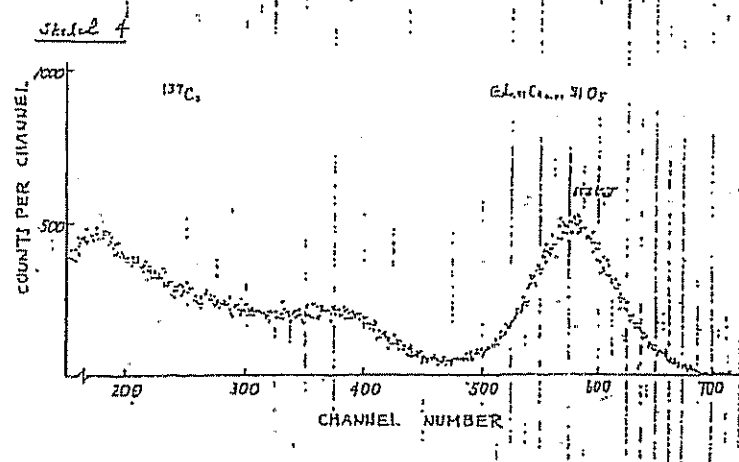
Lights having passed through the double slits were directed to be incident on a photomultiplier tube (PMT). The photomultiplier tube used was R878 available from Hamamatsu Photonics, Hamamatsu, Japan, which is known to have a photosensitivity as shown in Sketch 2. The peak wavelength of emission was determined from the measured spectrum.

Gamma ray spectra were measured in the system as shown in Sketch 3. Gamma ray source was accommodated in a lead container and placed on a shield covering the scintillator. The scintillator crystal was optically coupled to a photomultiplier tube (Hamamatsu Photonics R878) through optical grease. The photomultiplier tube was applied with a high tension voltage of 1000V and supplied the output signal. The output signal was amplified by an amplifier (ORTEC 113, time constant  $\tau=50$   $\mu$ sec, input capacitor  $C=100$  pF) and detected by multi-channel analyzer (MCA Techtronics 465).

#### Results:

The emission peak wavelength of  $Gd_{1.99}Ce_{0.01}SiO_3$  was found to be 430 nm.

The scintillation response spectra of  $Gd_{1.99}Ce_{0.01}SiO_3$  scintillator for  $^{137}Cs$  and  $^{60}Co$  measured by the



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aforementioned measuring system are as shown in Sketches 4 and 5.

It was confirmed that a scintillator crystal of  $Gd_{1.99}Ce_{0.01}SiO_5$  was practically useful as a scintillator crystal in a gamma ray detector.

5. One emission structure may be responsible to several kinds of excitation. There may be difference in the peak wavelength of emission by different excitation probably due to the difference in the nature of excitation.

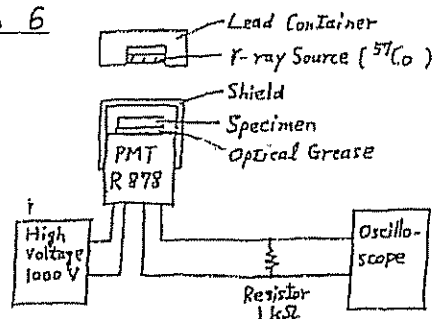
Gamma rays, especially intense  $\gamma$  rays, are hazardous to the health of human beings. Use of intense  $\gamma$  rays is dangerous for human beings except for in specially designed instruments. Therefore, it is usually difficult to measure  $\gamma$ -ray-excited emission spectra and to determine the peak wavelength of  $\gamma$ -ray-excited emission.

It has been empirically observed, however, that the difference in the peak wavelength of emission cannot be significantly large and is at most about 10 nm between EB excitation and  $\gamma$  ray excitation.

6. I examined the following four materials:

- (1)  $Y_{2.995}Ce_{0.015}Al_5O_{12}$ ,
- (2)  $Gd_{1.995}Ce_{0.015}Al_5O_{12}$ ,
- (3)  $Y_{0.995}Ce_{0.005}Al_5O_{12}$ , and
- (4)  $Gd_{0.995}Ce_{0.005}Al_5O_{12}$ .

Sketch 6



Experimental:

Specimen single crystals were shaped and polished to optical grade.

Gamma ray response was measured in a system as shown in Sketch 6.

Excitation source was  $^{57}\text{Co}$  (122 KeV) and was placed 50 mm just above the sample. The intensity of the source was 100  $\mu\text{C}$  when purchased and was estimated to be below 30  $\mu\text{C}$  when used in the experiment based on the half life of 270 days. Emission was detected by a photomultiplier tube (PMT) R-978 Hamamatsu Photonics applied with 1000V. The current output of the PMT was converted into a voltage by 1 k $\Omega$  resistor. The obtained readings in mV units were recorded as Relative Intensity.

Emission peak wavelength was measured under EB excitation in the system of Sketch 1 described in the above paragraph 4.

Results:

Experimental results are summarized in the following table.

- 8 -

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Sample No.	Compound	Relative Intensity	Emission peak wavelength
1	$\text{Y}_{2.995}\text{Ce}_{0.015}\text{Al}_5\text{O}_{12}$	10	550 nm
2	$\text{Gd}_{2.995}\text{Ce}_{0.015}\text{Al}_5\text{O}_{12}$	0	-
3	$\text{Y}_0.995\text{Ce}_{0.005}\text{AlO}_3$	3-4	370 nm
4	$\text{Gd}_{0.995}\text{Ce}_{0.005}\text{AlO}_3$	0	-

Experimental data showed that compound 1 exhibited scintillation under  $\gamma$  ray irradiation but compound 2 which differs from compound 1 in the point that Y is substituted by Gd did not exhibit scintillation under  $\gamma$  ray irradiation. Compound 3 is a known scintillator compound for X ray irradiation and also exhibited scintillation under  $\gamma$  ray irradiation. But, compound 4 which differs from compound 3 in the point that Y is substituted by Gd did not show scintillation under  $\gamma$  ray irradiation.

#### Analysis and Conclusion:

The sensitivity curve of the used photomultiplier covers almost the whole wavelength range of visible light. There were found considerable emission from the samples (1) and (3) but no recognizable emission from the samples (2) and (4).

Therefore, even when a compound containing Y does serve as a scintillator for  $\gamma$  ray irradiation, it cannot be estimated whether or not another compound in which Y is substituted with Gd can also serve as a scintillator for  $\gamma$  ray irradiation.

The undersigned declares further that all statements made herein of his own knowledge is true and that all statements made on information and belief is believed to be true; and further that these statements were made with the knowledge that willful false statements and the like so made are punishable by fine or imprisonment, or both, under section 1001 of Title 18 <sup>Dec. 24, 1984</sup> *KT* of the United States Code and that such willful false statements may jeopardize the validity of the application or any patent issuing thereon.

Dec. 24, 1984  
(Date)

Kazumasa Takagi  
KAZUMASA TAKAGI



224  
AZ 115-I - 539/15732

PATENT

160-120 001

IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

Applicant : Charles L. Melcher  
Serial No.: 07/389,502 - 4 Group Art Unit: 256  
Filed : August 4, 1989  
For : LUTETIUM ORTHOSILICATE  
SINGLE CRYSTAL  
SCINTILLATOR DETECTOR

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FBI - NEW YORK  
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PETITION FOR FOREIGN FILING LICENSE

Hon. Commissioner of Patents and Trademarks  
Washington, D.C. 20231

Attention: Licensing and Review Section

Expedited Handling Requested

Sir:

Applicant hereby petitions pursuant to 37 C.F.R. § 5.12(b) for a foreign filing license for the above-captioned patent application under 35 U.S.C. § 104. Applicant requests expedited handling of this petition, and that the foreign filing license be delivered to Applicant's attorneys at the address set forth at the end of this petition. A check for the required fee of \$120.00 pursuant to 37 C.F.R. § 1.07(h) is herewith enclosed. Expedited handling of this petition is respectfully requested in view of the fact that Applicant must file foreign patent applications on or before October 6, 1989 in order to claim the priority date of U.S. Patent Application Serial No. 07/254,353, which is the parent of the above-captioned U.S. patent application.

SMS003240

A2 J15-I - 539/15732

The present patent application is a continuing application of previously filed U.S. Patent Application Serial No. 07/254,353, filed October 5, 1988, which was granted a foreign filing license on October 27, 1988 (see Exhibit A attached hereto). The present patent application differs from its parent in that it adds to the parent disclosure a description of a typical logging tool in which the lutetium orthosilicate single crystal scintillator detector disclosed in the parent may be used and a drawing figure illustrating the tool. This description and drawing are essentially identical to disclosure material included in co-assigned U.S. Patent Application Serial No. 07/149,953, filed February 2, 1988 and allowed April 25, 1989, which is a continuing application of co-assigned U.S. Patent Application Serial No. 06/812,220, filed December 23, 1985. U.S. Patent Application Serial No. 07/149,953 is discussed at page 3, lines 21-25 of the specification of the above-captioned patent application.

Although a foreign filing license was not expressly granted in the filing receipt for U.S. Patent Application Serial No. 07/149,953 (see Exhibit B attached hereto), it is respectfully submitted that the subject matter of U.S. Patent Application Serial No. 07/149,953 requires no license for foreign filing in view of the fact that at least six months has passed since the February 2, 1988 filing date of the subject application with no secrecy order having been issued (see 37 C.F.R. § 5.11(e)(2)).

The present patent application discloses subject matter, all of which is disclosed either in its parent U.S. Patent Application Serial No. 07/254,353, which was

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previously granted a foreign filing license, or co-assigned U.S. Patent Application Serial No. 07/149,953, for which a foreign filing license is not required pursuant to 37 C.F.R. § 5.11(e)(2). Thus, it is respectfully submitted that the granting of the requested license will not result in any new matter being exported abroad for foreign filing.

In view of the foregoing, request is hereby made under 35 U.S.C. § 184 for a license to file foreign applications substantially identical with the patent disclosure of the above-captioned patent application. Such foreign filing is not considered likely to be detrimental to the public safety or defense and does not involve any secrecy order or national security of the United States.

Please deliver the foreign filing license requested herein to the following address:

Louis S. Sorell, Esq.  
Brumbaugh, Graves, Donohue & Raymond  
c/o Millen, White & Zelano, P.C.  
2200 Clarendon Boulevard  
Arlington, Virginia 22201

Respectfully submitted,



Arthur S. Tenser  
Patent Office Reg. No. 18,039

Richard G. Berkley  
Patent Office Reg. No. 25,465

Louis S. Sorell  
Patent Office Reg. No. 12,439

Attorneys for Applicant  
(212)408-2620

Enclosure



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11-3-89  
CR-4

27515-1 539/15132  
PATENT

GP 256  
#5  
2/24/90

IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

Applicants: Charles L. Melcher  
Serial No.: 07/389,502  
Filed : August 4, 1989 Group Art Unit:  
For : LUTETIUM ORTHOSILICATE  
SINGLE CRYSTAL  
SCINTILLATOR DETECTOR

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OFFICE: CHURCH 250

November 3, 1989

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NOV 15 1989  
LICENSING & REVIEW

I hereby certify that this paper is being deposited with the United States Postal Service as first class mail in an envelope addressed to: Commissioner of Patents and Trademarks, Washington, D.C. 20231, on

November 3, 1989  
Date of Deposit

Arthur S. Tenser 18,839  
Attorney Name Registration No.

*Arthur S. Tenser* November 3, 1989  
Signature Date of Signature

INFORMATION DISCLOSURE STATEMENT

Hon. Commissioner of Patents and Trademarks  
Washington, D.C. 20231

Sir:

Pursuant to 37 C.F.R. §§1.97 and 1.98, applicant resubmits the reference information incorporated in the Remarks to the amendment dated February 24, 1989 in parent application Serial No. 07/254,353 (now abandoned), and the form PTO-1449 submitted therewith. Copies of the listed references were included with the submission in the parent application.

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27515-1 539/15132  
PATENT

Buisson et al. disclose the crystallographic parameters of lutetium oxyorthosilicate powder along with those for other rare earth oxyorthosilicates.

Gomes de Mesquita et al. disclose cerium-activated lutetium oxyorthosilicate powders for use as cathode-ray phosphors.

Anan'eva et al. describe the growth by the Czochralski method of undoped single crystals of oxyorthosilicates of lanthanides from gadolinium to lutetium, and of yttrium oxyorthosilicate.

Holse et al. describe the optical properties of  $\text{Eu}^{3+}$ -activated rare earth oxyorthosilicate powders, wherein the rare earth elements were gadolinium, yttrium and lutetium.

Takagi et al. relates to the same general subject matter of the Takagi et al. patent already of record, i.e., a cerium-activated gadolinium oxyorthosilicate single crystal for use in position computed tomography.

Brandle et al. describe the growth of crystals of the type  $\text{Ln}_2\text{SiO}_5$  (Ln is a lanthanide) by the Czochralski method. Of the four original lanthanides selected for evaluation ( $\text{La}_2\text{SiO}_5$ ,  $\text{Ce}_2\text{SiO}_5$ ,  $\text{Gd}_2\text{SiO}_5$ , and  $\text{Y}_2\text{SiO}_5$ ) only two ( $\text{Gd}_2\text{SiO}_5$  and  $\text{Y}_2\text{SiO}_5$ ) were found suitable for Czochralski growth experiments.

Rabinovich et al. disclose the preparation of sols and gels of  $\text{Tb}^{+3}$ -doped yttrium orthosilicate for use as cathodoluminescent films.

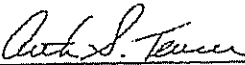
Blasse et al. describe the luminescence properties of a large number of phosphors or phosphor families. The authors conclude that while much is known about phosphors,

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PATENT

much is not well known and is subject to empirical  
prediction, if at all.

Ishii et al. (English language) and the last listed  
Japanese-language paper (of which a translation is not  
available) both appear to relate to the cerium-activated GSO  
crystal scintillator of the Takagi patent of record. They  
are being submitted for completeness.

Respectfully submitted,

  
Arthur S. Tonsor  
Reg. No. 18,839  
Attorney for Applicant  
(212) 408-2542

Enclosures





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Sheet 2 of 4

INFORMATION DISCLOSURE STATEMENT  
BY APPLICANT  
(Use several sheets if necessary)

Atty. Docket No. 27515-539/11889	Serial No. 07/254,353
Applicant	

Applicant  
Charles L. Melchor

Piling Date:	Group
October 6, 1988	113

## U.S. PATENT DOCUMENTS

[illegible]

FOREIGN PATENT DOCUMENTS

[illegible]

OTHER DOCUMENTS (including Author, Title Date, Pertinent Pages, Etc.)

J		<p>"SITE SELECTIVELY EXCITED LUMINESCENCE OF <math>\text{Eu}^{3+}</math> IN GADOLINIUM, YTTRIUM AND LUTETIUM OXYORTHOSILICATES" Nolsa et al., Journal of the Less-Common Metals, 126 (1986) 215-220.</p>
J		<p>"CERIUM-ACTIVATED <math>\text{Gd}_2\text{SiO}_5</math> SINGLE CRYSTAL SCINTILLATOR", Takagi et al., Appl. Phys. Lett., Vol. 42, No. 1, 1 January 1981 pp. 43-45.</p>
J		<p>"CZOCZRALSKI GROWTH OF RARE-EARTH ORTHOSILICATES (<math>\text{Ln}_2\text{SiO}_5</math>)", Brandle et al., Journal of Crystal Growth 79 (1986) pp. 308-315.</p>

**Examiner**

T Corp

Date Considered
-----------------

2/20/40

<sup>A</sup>Examiner: Initial citation considered, whether or not citation is in conformance with HPEP 609; Draw line through citation if not in conformance and not considered. Include copy of this form with next communication to applicant.

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Sheet 4 of 4

FORM 100-1449  
(REV. 2-82)

U.S. Department of Commerce  
Patent and Trademark Office

Atty. Docket No.  
27575-539/11889

Serial No.  
07/254,353

Applicant  
Charles L. Melcher

INFORMATION DISCLOSURE STATEMENT  
BY APPLICANT  
(Use several sheets if necessary)

Filing Date  
October 6, 1988

Group  
113

## U.S. PATENT DOCUMENTS

[illegible]

## FOREIGN PATENT DOCUMENTS

[illegible]

OTHER DOCUMENTS (Including Author, Title Date, Pertinent Pages, Etc.)

	"Ce-Cd <sub>2</sub> SiO <sub>5</sub> " (Japanese language)

Examiner

J. Cooper

Date Considered

2 | 20 Ki

\*Examiner: Initial citation considered, whether or not citation is in conformance with MPEP 609; draw line through citation if not in conformance and not considered. Include copy of this form with next communication to applicant.

SMS003249



UNITED STATES DEPARTMENT OF COMMERCE  
Patent and Trademark Office  
Address: COMMISSIONER OF PATENTS AND TRADEMARKS  
Washington, D.C. 20231

SERIAL NUMBER	FILING DATE	FIRST NAMED APPLICANT	ATTORNEY DOCKET NO.
146812			

EXAMINER	
ART UNIT	PAPER NUMBER
	6

DATE MAILED:

## EXAMINER INTERVIEW SUMMARY RECORD

All participants (applicant, applicant's representative, PTO personnel):

(1) Mr. Berkley (2) \_\_\_\_\_

(3) \_\_\_\_\_ (4) \_\_\_\_\_

Date of interview: 5/2/89Type: ☒ Telephonic ☐ Personal (copy if given to ☐ applicant ☐ applicant's representative).Exhibit shown or demonstration conducted: ☐ Yes ☐ No. If yes, brief description: \_\_\_\_\_Agreement ☒ was reached with respect to some or all of the claims in question. ☐ was not reached.Claims discussed: claim 2

Identification of prior art discussed: \_\_\_\_\_

Description of the general nature of what was said to if an agreement was reached, or any other comments:

See Examiner's Amendment

IA fuller description, if necessary, and a copy of the amendment, if available, which the examiner agreed would render the claims allowable must be attached. Also, where no copy of the amendment which would render the claims allowable is available, a summary thereof must be attached.

Under the paragraphs below have been checked to indicate to the contrary, A FORMAL WRITTEN RESPONSE TO THE LAST OFFICE ACTION IS NOT WAIVED AND MUST INCLUDE THE SUBSTANCE OF THE INTERVIEW (e.g., items 1-7 on the reverse side of this form). If a response to the last Office action has already been filed, then applicant is given one month from this interview date to provide a statement of the substance of the interview.

☒ It is not necessary for applicant to provide a separate record of the substance of the interview.

☐ Since the examiner's interview summary above (including any attachments) reflects a complete response to each of the objections, rejections and requirements that may be present in the last Office action, and since the claims are now allowable, this completed form is considered to fulfill the response requirements of the last Office action.

PTOL-413 (REV. 1-84)

APPLICANT'S COPY

Examiner's Signature

SMS003250



UNITED STATES DEPARTMENT OF COMMERCE  
Patent and Trademark Office  
Address: COMMISSIONER OF PATENTS AND TRADEMARKS  
Washington, D.C. 20231

SERIAL NUMBER	FILING DATE	FIRST NAMED APPLICANT	ATTORNEY DOCKET NO.
389502			

EXAMINER	
ART UNIT	PAPER NUMBER

DATE MAILED

## EXAMINER INTERVIEW SUMMARY RECORD

All participants (applicant, applicant's representative, PTO personnel):

(1) Mr. Tencer (3) \_\_\_\_\_  
(2) \_\_\_\_\_ (4) \_\_\_\_\_

Date of interview 7/20/00Type: ☒ Telephonic ☐ Personal (copy is given to ☐ applicant ☐ applicant's representative).Exhibit shown or demonstration conducted: ☐ Yes ☐ No. If yes, brief description: \_\_\_\_\_Agreement ☒ was reached with respect to some or all of the claims in question. ☐ was not reached.

Claims discussed: \_\_\_\_\_

Identification of prior art discussed: \_\_\_\_\_

Description of the general nature of what was agreed to if an agreement was reached, or any other comments: \_\_\_\_\_

See Examiner's Amendment

[A fuller description, if necessary, and a copy of the amendments, if available, which the examiner agreed would render the claims allowable must be attached. Also, where no copy of the amendments which would render the claims allowable is available, a summary thereof must be attached.]

Unless the paragraphs below have been checked to indicate to the contrary, A FORMAL WRITTEN RESPONSE TO THE LAST OFFICE ACTION IS NOT WAIVED AND MUST INCLUDE THE SUBSTANCE OF THE INTERVIEW (e.g., Items 1-7 on the reverse side of this form). If a response to the last Office action has already been filed, then applicant is given one month from this interview date to provide a statement of the substance of the interview.

☒ It is not necessary for applicant to provide a separate record of the substance of the interview.☐ Since the examiner's interview summary above (including any attachments) reflects a complete response to each of the objections, rejections and requirements that may be present in the last Office action, and since the claims are now allowable, this completed form is considered to fulfill the response requirements of the last Office action.

Examiner's Signature

PTO-813 (REV. 1-94)

ORIGINAL FOR INSERTION IN TIGHT RING FLAP OF FILE WRAPPER

SMS003251



UNITED STATES DEPARTMENT OF COMMERCE  
Patent and Trademark Office  
Address: COMMISSIONER OF PATENTS AND TRADEMARKS  
Washington, D.C. 20231

SERIAL NUMBER 11773671-01	FILING DATE 03/01/90	FIRST NAMED APPLICANT RELCORP	ATTORNEY DOCKET NO. 215751631171
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BRUNBAUCH, GRAVES, DOMOHUE & RAYMOND  
36 ROCKEFELLER PLAZA  
NEW YORK, NY 10112

COOPER, J	PAPER NUMBER
-----------	--------------

ART UNIT 113  
DATE MAILED: 03/01/90

### NOTICE OF ALLOWABILITY

#### PART I.

- 1 ☐ This communication is responsive to \_\_\_\_\_
- 2 ☒ As the claims being allowable, PROSECUTION ON THE MERITS IS (OR REMAINS) CLOSED in this application. If not included herewith (or previously mailed), a Notice Of Allowance And Issue Fee Due of other appropriate communication will be sent in due course.
- 3 ☒ The allowed claims are 3 and 4
- 4 ☐ The drawings filed on \_\_\_\_\_ are acceptable.
- 5 ☐ Acknowledgment is made of the claim for priority under 35 U.S.C. 119. The certified copy has ( ) been received ( ) not been received ( ) been filed in parent application Serial No. \_\_\_\_\_ filed on \_\_\_\_\_
- 6 ☒ Note the attached Examiner's Amendment
- 7 ☒ Note the attached Examiner Interview Summary Record, PTO-413
- 8 ☐ Note the attached Examiner's Statement of Reasons for Allowance
- 9 ☒ Note the attached NOTICE OF REFERENCES CITED, PTO-892
- 10 ☒ Note the attached INFORMATION DISCLOSURE CITATION, PTO-1449.

#### PART II.

A SHORTENED STATUTORY PERIOD FOR RESPONSE to comply with the requirements noted below is set to EXPIRE THREE MONTHS FROM THE "DATE MAILED" indicated on this form. Failure to timely comply will result in the ABANDONMENT of this application. Extensions of time may be obtained under the provisions of 37 CFR 1.136(a).

- 1 ☐ Note the attached EXAMINER'S AMENDMENT or NOTICE OF INFORMAL APPLICATION, PTO-152, which discloses that the oath or declaration is deficient. A SUBSTITUTE OATH OR DECLARATION IS REQUIRED.
- 2 ☒ APPLICANT MUST MAKE THE DRAWING CHANGES INDICATED BELOW IN THE MANNER SET FORTH ON THE REVERSE SIDE OF THIS PAPER.
  - a ☐ Drawing Informalities are indicated on the NOTICE RE PATENT DRAWINGS, PTO-948, attached hereto or to Paper No. \_\_\_\_\_. CORRECTION IS REQUIRED.
  - b ☐ The proposed drawing correction filed on \_\_\_\_\_ has been approved by the examiner. CORRECTION IS REQUIRED.
  - c ☐ Approved drawing corrections are described by the examiner in the attached EXAMINER'S AMENDMENT. CORRECTION IS REQUIRED.
  - d ☒ Formal drawings are now REQUIRED.

Any response to this letter should include in the upper right hand corner, the following information from the NOTICE OF ALLOWANCE AND ISSUE FEE DUE: ISSUE DATA NUMBER, DATE OF THE NOTICE OF ALLOWANCE, AND SERIAL NUMBER.

#### Attachments:

- |  |   |
|--|---|
| - Examiner's Amendment                       | - Notice of Informal Application, PTO-152 |
| - Examiner Interview Summary Record, PTO-413 | - Notice to Patent Drawings, PTO-948      |
| - Reasons for Allowance                      | - Listing of Docket Drawings              |
| - Notice of References Cited, PTO-892        | - Oath                                    |
| - Information Disclosure Citation, PTO-1449  |   |

PTOL-27 (REV. 11-81)

*Jack Cooper*  
JACK COOPER  
PRIMARY PATENT EXAMINER  
ART UNIT 113

SMS003252

Serial No. 389,502

-2-

Art Unit 113

Examiner's Amendment

Restriction to one of the following inventions is required under 35 U.S.C. 121:

I. Claims 1-4, drawn to a scintillator and detector, classified in Class 250, subclass 483.1.

II. Claims 5-12, drawn to an apparatus, classified in Class 250, subclass 269.

The inventions are distinct, each from the other, because of the following reasons:

Inventions I and II are related as combination and subcombination. Inventions in this relationship are distinct if it can be shown that (1) the combination as claimed does not require the particulars of the subcombination as claimed for patentability, and (2) that the subcombination has utility by itself or in other combinations. (MPEP 806.05(c)).

In this case, the combination as claimed does not require the particulars of the subcombination as claimed because the lutetium ~~silicate~~ oxyorthosilicate scintillator: (1) does not have to have the formulation of the scintillator of Group I, (2) does not have to be in transparent single crystal form, and (3) does not have to have a photodetector coupled thereto -see claim 5. The subcombination has separate utility such as a detector in nuclear medicine.

Because these inventions are distinct for the reasons given above, and have acquired a separate status in the art as shown by their different classification restriction for examination purposes as indicated is proper.

---

SMS003253

Serial No. 389,502  
Art Unit 113

-3-

During a telephone conversation with Mr. Tenser, applicant's counsel of record on February 20, 1990 a provisional election was made with traverse to prosecute the invention of Group I, claims 1-4. Affirmation of this election must be made by applicant in responding to this Office action, and such affirmation must distinctly and specifically point out the reasons upon which applicant bases his or her conclusion that the requirement to restrict is in error. Claims 5-12 are withdrawn from further consideration by the Examiner, 37 CFR 1.142(b), as being drawn to a nonselected invention.

Group I claims were examined and found to contain patentable subject matter. Mr. Tenser was informed of this finding in a telephone interview on February 20, 1990 and authorized the Examiner to make the following changes:

Page 1, line 5 --in part-- has been inserted after "continuation", line 6 --, now abandoned-- has been inserted after "1988".

~~Page 9, line 23 --now U.S. Patent No. 4,891,956--~~  
~~has been inserted after "1988".~~

Claims 1, 2 and 5-12 have been cancelled.

J. Cooper:mn  
703-557-2517  
2-26-90

*J. Cooper*  
JACK COOPER  
PRIMARY PATENT EXAMINER  
ART UNIT 113

SMS003254

TO SEPARATE HOLD TOP AND BOTTOM EDGES SHAP-APART AND RE-SAND/CARDON

FORM PTO-892 (REV. 3-79)		U.S. DEPARTMENT OF COMMERCE PATENT AND TRADEMARK OFFICE		SERIAL NO. 389502	FIGURE UNIT 113	ATTACHMENT TO PAPER NUMBER		
NOTICE OF REFERENCES CITED				APPLICANT'S Melcher				
U.S. PATENT DOCUMENTS								
		DOCUMENT NO.	DATE	NAME	CLASS	SUB-CLASS	FILING DATE IF APPROPRIATE	
✓	A	4208611	6/80	Watanabe et al	252	301.4F		
✓	B	4647781	3/87	Takagi et al	252	301.4F		
	C							
	D							
	E							
	F							
	G							
	H							
	I							
	J							
	K							
FOREIGN PATENT DOCUMENTS								
		DOCUMENT NO.	DATE	COUNTRY	NAME	CLASS	SUB-CLASS	PERTINENT SHEETS OF PAGES
✓	L	5159079	5/76	Japan	Matsumura	252	301.4F	
	M							
	N							
	O							
	P							
	Q							
OTHER REFERENCES (Including Author, Title, Date, Pertinent Pages, Etc.)								
	R							
	S							
	T							
	U							
EXAMINER J. Cooper				DATE 2/20/90				
* A copy of this reference is not being furnished with this office action. (See Manual of Patent Examining Procedure, section 707.05 (d).)								

SMS003255



2...15-1 539/15132  
PATENT

## IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

Group 116:

Applicant: Charles L. Melcher  
 Serial No.: 07/389,502-4 Examiner: Cooper  
 Filed : August 4, 1989 Group Art Unit: 256  
 For : LUTETIUM ORTHOSILICATE SINGLE  
 CRYSTAL SCINTILLATOR DETECTOR

RECEIVED

APR 12 1990

L E T T E R

RECEIVED

MAY 25 1990

LICENSING &amp; REVIEW

LICENSING &amp; REVIEW

I hereby certify that this paper was being deposited with the United States Postal Service as first class mail in an envelope addressed to: Commissioner of Patents and Trademarks, Washington, D.C. 20231; on

April 16, 1990  
 Date of Deposit

Arthur S. Tenser  
 Attorney Name

18,839  
 Registration No.

*Arthur S. Tenser*  
 Signature

April 16, 1990  
 Date of Signature

Hon. Commissioner of Patents and Trademarks  
 Washington, D.C. 20231

Sir:

This letter is responsive to the Notice of Allowability and accompanying Examiner's Amendment and Examiner's Interview Summary Record dated March 1, 1990.

As required, formal drawings are submitted herewith.

The applicant affirms the election of the invention of Group 1, claims 1-4, made in response to the requirement for restriction noted in the Examiner's Amendment. This election was made with traverse during the course of the


SMS003256

2,15-1 539/15132  
PATENT

telephone conversation on February 20, 1990, as noted by the Examiner. Inasmuch as it was agreed to cancel non-elected claims to expedite prosecution of the application, further argument as to the merits of the requirement for restriction is deemed unnecessary.

In view of the Examiner's Amendment and the submission of formal drawings herewith, the application is believed in condition for immediate allowance and action to that effect is respectfully solicited.

Respectfully submitted,

  
Arthur S. Tenser  
Reg. No. 18,839

Attorney for Applicant  
(212) 408-2542

Enclosures

---

SMS003257

TO-1000 (Rev. 5-79) RECEIPT FILE		MAY 02 1990 381502		MERCE #8 TRADEMARK
SERIAL NUM.		GROUP NO.	JUDICIAL NO.	DOCKET NO.
EXAMINED BY		AUG 14 89 07 970689		
APPLICANT		INVENTOR		
DATE		INVENTION		
DATES FIRST SECOND THIRD FOURTH FIFTH		DESIGNS TOTAL LVS	DESIGNS TOTAL LVS	DESIGNS TOTAL LVS
DOE		DOE		

Title 35, United States Code, Section 181

I hereby acknowledge that I have inspected the disclosure of the above identified application for patent in the administration of the law cited above, on behalf of the department or agency which I represent, and promise that any information acquired from said application will not be divulged, disclosed or used for any purpose other than in the administration of the cited law.

NAME	DATE	AGENCY REPRESENTED
V. Caron	4-27-90	DOE

SMS003258

FTO-1031 (Rev. 8-78) SECURITY FILE		GROUP NO. <u>1289502</u> GROUP DATE <u>AUG 14 89 07</u> SERIAL NO. <u>9N689</u> NUMBER OF PAGES	
EXEMPT BY NONE <input type="checkbox"/> DATE DARCOM <input type="checkbox"/> NAVY <input type="checkbox"/> AF <input type="checkbox"/> CE <input type="checkbox"/> DOE <input type="checkbox"/> NASA <input type="checkbox"/> NSA <input type="checkbox"/>		MATICARD DIVISION CLASSIFIED TOTAL PAGES LOG COPY TRANSFER FILE TRANSLATION AUTH. CL.	
CLEARED BY GROUP 220, SECURITY		RECOMMENDATION BY EXPERTS (Every expert examining this application should indicate an express RECOMMENDATION followed by their SIGNATURE, AGENCY AND DATE)	
<div style="display: flex; justify-content: space-around; align-items: center;"> <div> <input type="checkbox"/> DARCOM NAVY         </div> <div> <input type="checkbox"/> AF AF         </div> <div> <input type="checkbox"/> CE CE         </div> <div> <input type="checkbox"/> DOE DOE         </div> <div> <input type="checkbox"/> NASA NASA         </div> <div> <input type="checkbox"/> NSA NSA         </div> <div> <input type="checkbox"/> LOG LOG         </div> <div> <input type="checkbox"/> 185 185         </div> <div> <input type="checkbox"/> OTHER OTHER         </div> </div>			

SMS003259

TO-100C REV. 5-78 SECURITY ID#  SEARCHED BY  INDEXED <input type="checkbox"/> DATE  DALLAS MIAMI ALBANY ALBUQUERQUE ANCHORAGE ARIZONA BIRMINGHAM BUTTE CHARLOTTE CHICAGO CINCINNATI COVINGTON DENVER DETROIT EL PASO HOUSTON INDIANAPOLIS JACKSONVILLE JAXSONVILLE KANSAS CITY LITTLE ROCK LOS ANGELES MEMPHIS MILWAUKEE MINNEAPOLIS MOBILE NEW ORLEANS NEW YORK OMAHA OKLAHOMA CITY PHOENIX PORTLAND RICHMOND SACRAMENTO SAN ANTONIO SAN DIEGO SAN JOSE SEATTLE SFO TAMPA TULSA WASHINGTON FIELD WASHINGTON METRO WASHINGTON STATE WASHINGTON TERRITORY YAKIMA	CASE NO. AUG 14 89 07 ATTORNEY REFERENCE	389502 INITIALS AND NUMBER OF PAGES 4 MV689	
	SEARCHED TOTAL (S) IND CT FRING THE ETC TRANSFERRED ASSTY DL		
	ACCESS ACKNOWLEDGEMENT As Required by Title 35, United States Code (1952) Section 101		
	I hereby acknowledge that I have inspected the disclosure of the above identified application for patent, in the administration of the law cited above, on behalf of the department or agency which I represent, and promise that any information acquired from said application will not be divulged, disclosed or used for any purpose other than in the administration of the cited law.		
NAME	DATE	AGENCY REPRESENTED	

SMS003260



UNITED STATES DEPARTMENT OF COMMERCE  
Patent and Trademark Office

Address: Box ISSUE FEE  
COMMISSIONER OF PATENTS AND TRADEMARKS  
Washington, D.C. 20231

BRUMBAUGH, GRAVES, DONOHUE & RAYMOND  
30 ROCKEFELLER PLAZA  
NEW YORK, NY 10112

**NOTICE OF ALLOWANCE  
AND ISSUE FEE DUE**

- ☐ This notice is being sent to you by the Examiner.  
☐ This notice is being sent to you by the Commissioner of Patents and Trademarks.

SERIES/CONVENTION NO.	FILING DATE	TOTAL CLAIMS	EXAMINER AND DISCUSSION UNIT	DATE MAILED
07/389,502	08/04/89	002	COOPER, J	117 05/08/90
Inventor(s) Applicant	MELCHER,	CHARLES L.		

TITLE OF INVENTION: LUTETIUM ORTHOSILICATE SINGLE CRYSTAL SCINTILLATOR DETECTOR

ATTY DOCKET NO.	CLASS/CLASSICAL	DATCH NO.	APPL. TYPE	SMALL ENTITY	FEE DUE	DATE DUE
I 275191539151	250-483.100	F33	UTILITY	NO	\$620.00	08/08/90

**THE APPLICATION IDENTIFIED ABOVE HAS BEEN EXAMINED AND IS ALLOWED FOR ISSUANCE AS A PATENT.  
PROSECUTION ON THE MERITS IS CLOSED.**

**THE ISSUE FEE MUST BE PAID WITHIN THREE MONTHS FROM THE MAILING DATE OF THIS NOTICE OR THIS  
APPLICATION SHALL BE REGARDED AS ABANDONED. THIS STATUTORY PERIOD CANNOT BE EXTENDED.**

**HOW TO RESPOND TO THIS NOTICE:**

**I. Review the SMALL ENTITY Status shown above.**

If the SMALL ENTITY is shown as YES, verify your current SMALL ENTITY status:

- A. If the Status is changed, pay twice the amount of the FEE DUE shown above and notify the Patent and Trademark Office of the change in status, or  
B. If the Status is the same, pay the FEE DUE shown above.

If the SMALL ENTITY is shown as NO:

- A. Pay FEE DUE shown above, or  
B. File verified statement of Small Entity Status before, or with payment of 1/2 the FEE DUE shown above

II. Part B of this notice should be completed and returned to the Patent and Trademark Office (PTO) with your ISSUE FEE. Even if the ISSUE FEE has already been paid by a charge to deposit account, Part B should be completed and returned. If you are charging the ISSUE FEE to your deposit account, Part C of this notice should also be completed and returned.

III. All communications regarding this application must give serial code (or filing date), serial number and batch number. Please direct all communications prior to issuance to Box ISSUE FEE unless advised to the contrary.

**IMPORTANT REMINDER: Patents Issued on applications filed on or after Dec. 12, 1980 may require payment of maintenance fees.**

PTOL-35 (REV. 12-89) (2000) (2000) (2000)

PATENT AND TRADEMARK OFFICE COPY

SMS003261



UNITED STATES DEPARTMENT OF COMMERCE  
Patent and Trademark Office  
Address: COMMISSIONER OF PATENTS AND TRADEMARKS  
Washington, D.C. 20231

SERIAL NUMBER	FILING DATE	FIRST NAMED APPLICANT	ATTORNEY DOCKET NO.
07/389,502	08/04/89	CHARLES L. MELCHER	

OF RECORD:

EXAMINER	
ART UNIT	PAPER NUMBER
	10

DATE MAILED: May 16, 1990

A statement under Section 152 of Public Law 783 (83rd Congress, 2nd Session), concerning the making of the invention or discovery described therein, has been filed in the above-identified application. A copy of the application and a copy of the statement are forwarded herewith.

A statement of the date of receipt in the U.S. Department of Energy of the copy of the statement and the copy of the application is requested to determine the starting of the 90-day period recited in paragraph 2 of Section 152. An endorsed copy of this letter is provided for this purpose.

By direction of the Commissioner

*for Williams*  
Special Laws Administration Group

Encl: Photo statement  
Photo application  
Receipt copy of this letter

Receipt in the U.S. Department of Energy is acknowledged of a copy of the above letter and its enclosure.

Date  
THIS CORRESPONDENCE TO:

Assistant General Counsel  
for Patents, GC-42  
U.S. Department of Energy  
1000 Independence Ave.  
MS 6F-067  
Washington, D.C. 20585

Signed

FD-210 (Rev. 4-83)

SMS003262



UNITED STATES DEPARTMENT OF COMMERCE  
Patent and Trademark Office  
Address: COMMISSIONER OF PATENTS AND TRADEMARKS  
Washington, D.C. 20231

SERIAL NUMBER	FILING DATE	FIRST NAMED APPLICANT	ATTORNEY DOCKET NO.
07/309,502	08/04/89	CYNTHIA L. MELCHER	

OF RECORD:

EXAMINER	
ART UNIT	PAPER NUMBER
	10
DATE MAILED: May 16, 1990	

RECEIVED  
JUL 2 1990  
LICENSING & REVIEW

A statement under Section 152 of Public Law 703 (81st Congress, 2nd Session), concerning the making of the invention or discovery described therein, has been filed in the above-identified application. A copy of the application and a copy of the statement are forwarded herewith.

A statement of the date of receipt in the U.S. Department of Energy of the copy of the statement and the copy of the application is requested to determine the starting of the 90-day period recited in paragraph 2 of Section 152. An endorsed copy of this letter is provided for this purpose.

By direction of the Commissioner:

*[Signature]*  
Special Laws Administration Group

Encl: Photo statement  
Photo application  
Receipt copy of this letter

This application has been reviewed and from the information at hand you are hereby advised that the U.S. Department of Energy will not file a dispositive in this case.

Receipt in the U.S. Department of Energy is acknowledged of a copy of the above letter and its enclosure. JUN 13 1990

May 13 1990

Date

THIS CORRESPONDENCE TO:

Assistant General Counsel  
for Patents, GC-42  
U.S. Department of Energy  
1000 Independence Ave.  
MS 6F-067  
Washington, D.C. 20585

Signed

*[Signature]*  
Assistant General Counsel  
for Patents

PTO-215 (Rev. 4-89)

SMS003263



## PART B - ISSUE FEE TRANSMITTAL

**MAILING INSTRUCTIONS:** This form should be used for transmitting the ISSUE FEE. Blocks 2 through 6 should be completed where appropriate. All future correspondence including the Issue Fee Receipt, the Patent, advanced orders and notification of maintenance fees will be mailed to addressee entered in Block 1 unless you direct otherwise, by: (a) specifying a new correspondence address in Block 3 below; or (b) providing the PTO with a separate "FEE ADDRESS" for maintenance fee notifications with the payment of issue fee or duration. See reverse for Certificate of Mailing.

1. CORRESPONDENCE ADDRESS (If change from Block 1 of Form PTO/SF 100, enter new address)	2. INVENTOR(S) ADDRESS CHANGE (Complete only if there is a change)
BRUMBAUGH, GRAVES, DONOHUE & RAYMOND 30 ROCKEFELLER PLAZA NEW YORK, NY 10112	INVENTOR'S NAME Direct Address City, State and ZIP Code CO-INVENTOR'S NAME Direct Address City, State and ZIP Code <input type="checkbox"/> Check if additional changes are on reverse side

SEQUEL CODE/SERIAL NO.	PRIOR DATE	TOTAL CLAIMS	EXAMPER AND GROUP/ATTY UNIT	DATE PAID
07/089,502	08/03/89	002	CDOPER, J	117 05/08/90
Patentee/Applicant	NIELCHER, CHARLES L.			

TITLE OF INVENTION: **SEMICONDUCTOR SINGLE CRYSTAL SCINTILLATOR DETECTOR**

ATTY'S REGISTRY NO.	CLASS-DIV-CLASS	DATCH NO.	APPL. TYPE	SMALL ENTITY	FEE DUE	DATE DUE
1 275157039121	250-483.100	F99	UTILITY	NO	\$820.00	09/09/90

3. Further correspondence to be mailed to the following:	4. For printing on the patent front page, list the names of not more than 3 registered patent attorneys or agents OR alternatively, the name of a firm having as a member a registered attorney or agent. If no name is listed, no name will be printed.
	1. <u>Brumbaugh Graves</u> 2. <u>Donohue &amp; Raymond</u> 3. _____

0 HC 07/19/90 07309502

DO NOT USE THIS SPACE  
1 142 820.00 CK

PT11486 07/19/90 07309502 02-4377-110		The following fees are required:	
5. ASSIGNMENT DATA TO BE PRINTED ON THE PATENT (print or type)		<input checked="" type="checkbox"/> Issue Fee <input type="checkbox"/> Advanced Order - # of Copies _____ (Minimum of 10)	
(i) NAME OF ASSIGNEE: <b>Schlumberger Technology Corporation</b>		6b. The following fees should be charged to: DEPOSIT ACCOUNT NUMBER <b>02-4377</b> (Enclose Part 07)	
(ii) ADDRESS: (City & State or Country) <b>277 Park Avenue New York, NY 10172</b>		<input type="checkbox"/> Issue Fee <input checked="" type="checkbox"/> Advanced Order - # of Copies _____ (Minimum of 10)	
(iii) STATE OF INCORPORATION, IF ASSIGNEE IS A CORPORATION: <b>Texas</b>		<input checked="" type="checkbox"/> Any Delinquencies in Enclosed Fees	
A <input type="checkbox"/> This application is NOT assigned. <input checked="" type="checkbox"/> Assignment previously submitted to the Patent and Trademark Office. <input type="checkbox"/> Assignment is being submitted under corporate control. Assignments should be directed to Box ASSIGNMENTS.		THE COMMISSIONER OF PATENTS AND TRADEMARKS is requested to apply the Issue Fee to the application identified above. (Signature of person in receipt of record) <u>[Signature]</u> (Date) <b>7/12/90</b>	
PLEASE NOTE: Unless an assignment is identified in Block 5, no assignment data will appear on the patent. Inclusion of assignment data is only appropriate when an assignment has been previously submitted to the PTO or is being submitted under separate cover. Completion of this form is NOT a substitute for filing an assignment.		NOTE: The Issue Fee will not be accepted from anyone other than the applicant, a registered attorney or agent, or the assignee or other party in interest as shown by the records of the Patent and Trademark Office.	

TRANSMIT THIS FORM WITH FEE CERTIFICATE OF MAILING ON REVERSE

PTOL FEE (REV 12/90) Due when received

SMS003264

**Certificate of Mailing**

I hereby certify that this correspondence is being deposited with the United States Postal Service with sufficient postage as first class mail in an envelope addressed to:

Box ISSUE FEE  
Commissioner of Patents and Trademarks  
Washington, D.C. 20231

on July 12, 1990  
(Date)

Arthur E. Tesser  
(Name of person making deposit)

*Arthur E. Tesser*  
(Signature)

July 12, 1990  
(Date)

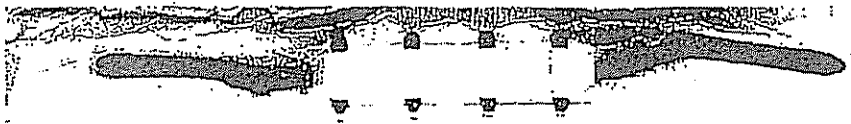
Note: If this certificate of mailing is used, it can only be used to transmit the Issue Fee. This certificate cannot be used for any other accompanying papers. Each additional paper, such as an assignment or formal drawings, must have its own certificate of mailing.

NO 00.000 511 1 5029050 09/21/90 3H  
100 011 11/1/90 5029050 09/21/90 4H11114

This form is estimated to take 20 minutes to complete. Time will vary depending upon the needs of the individual applicant. Any comments on the amount of time you require to complete this form should be sent to the Office of Management and Organization, Patent and Trademark Office, Washington, D.C. 20231 and to the Office of Information and Regulatory Affairs, Office of Management and Budget, Washington, D.C. 20503.

REVISED SEP 1984 (REV 11/85) (CUB) (Continued on page 2)

SMS003265



PTO UTILITY GRANT  
Paper Number 62

*The  
United  
States  
of  
America*


The Commissioner of Patents  
and Trademarks

*Has received an application for a patent  
for a new and useful invention. The title  
and description of the invention are en-  
closed. The requirements of law have  
been complied with, and it has been de-  
termined that a patent on the invention  
shall be granted under the law.*

Therefore, this

United States Patent

*Grants to the person or persons having  
title to this patent the right to exclude  
others from making, using or selling the  
invention throughout the United States  
of America for the term of seventeen  
years from the date of this patent, sub-  
ject to the payment of maintenance fees  
as provided by law.*



*Harry F. Marshall, Jr.*  
Commissioner of Patents and Trademarks

*Melvinia Gary*  
Agent

PTO-1584



IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

27515-1 539/15132  
PATENT

GR 256

920  
11/10/91

#13

Pat No 4958886 RECEIVED

DEC 06 1990

GROUP 250

Applicants: Charles L. Melcher  
Serial No.: 07/389,502  
Filed : August 4, 1989 Group Art Unit: 250  
For : LUTETIUM ORTHOSILICATE  
SINGLE CRYSTAL  
SCINTILLATOR DETECTOR

November 3, 1989

I hereby certify that this paper is being deposited with the United States Postal Service as first class mail in an envelope addressed to: Commissioner of Patents and Trademarks, Washington, D.C. 20231, on

November 3, 1989  
Date of Deposit

Arthur S. Tenser  
Attorney Name

18,879  
Registration No.

*Arthur S. Tenser*  
Signature

November 3, 1989  
Date of Signature

INFORMATION DISCLOSURE STATEMENT

Hon. Commissioner of Patents and Trademarks  
Washington, D.C. 20231

Sir:

Pursuant to 37 C.F.R. §§1.97 and 1.98, applicant resubmits the reference information incorporated in the Remarks to the amendment dated February 24, 1989 in parent application Serial No. 07/254,353 (now abandoned), and the form PTO-1449 submitted therewith. Copies of the listed references were included with the submission in the parent application.

SMS003267

2/515-1 539/15132  
PATENT

Buisson et al. disclose the crystallographic parameters of lutetium oxyorthosilicate powder along with those for other rare earth oxyorthosilicates.

Gomes de Mesquita et al. disclose cerium-activated lutetium oxyorthosilicate powders for use as cathode-ray phosphors.

Anan'eva et al. describe the growth by the Czochralski method of undoped single crystals of oxyorthosilicates of lanthanides from gadolinium to lutetium, and of yttrium oxyorthosilicate.

Holsa et al. describe the optical properties of  $\text{Eu}^{3+}$ -activated rare earth oxyorthosilicate powders, wherein the rare earth elements were gadolinium, yttrium and lutetium.

Takagi et al. relates to the same general subject matter of the Takagi et al. patent already of record, i.e., a cerium-activated gadolinium oxyorthosilicate single crystal for use in position computed tomography.

Brandle et al. describe the growth of crystals of the type  $\text{Ln}_2\text{SiO}_5$  (Ln is a lanthanide) by the Czochralski method. Of the four original lanthanides selected for evaluation ( $\text{La}_2\text{SiO}_5$ ,  $\text{Ce}_2\text{SiO}_5$ ,  $\text{Gd}_2\text{SiO}_5$ , and  $\text{Y}_2\text{SiO}_5$ ) only two ( $\text{Gd}_2\text{SiO}_5$  and  $\text{Y}_2\text{SiO}_5$ ) were found suitable for Czochralski growth experiments.

Rabinovich et al. disclose the preparation of sols and gels of  $\text{Tb}^{3+}$ -doped yttrium orthosilicate for use as cathodoluminescent films.

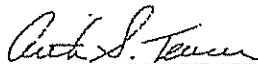
Blasse et al. describe the luminescence properties of a large number of phosphors or phosphor families. The authors conclude that while much is known about phosphors,

2,515-1 539/15132  
PATENT

much is not well known and is subject to empirical  
prediction, if at all.

Ishii et al. (English language) and the last listed  
Japanese-language paper (of which a translation is not  
available) both appear to relate to the cerium-activated GSO  
crystal scintillator of the Takagi patent of record. They  
are being submitted for completeness.

Respectfully submitted,

  
Arthur S. Tenser  
Reg. No. 18,839  
Attorney for Applicant  
(212) 408-2542

Enclosures



Sheet 1 of 4

Form PTO-1449 (REV. 2-87)	U.S. Department of Commerce Patent and Trademark Office	Atty. Docket No. 27515-539/11083	Serial No. 07/250,353
INFORMATION DISCLOSURE STATEMENT BY APPLICANT (Use several sheets if necessary)		Applicant Charles L. Mulicher	
		Filing Date October 6, 1988	Group 113

## U.S. PATENT DOCUMENTS

[illegible]

FOREIGN PATENT DOCUMENTS

[illegible]

OTHER DOCUMENTS (including Author, Title Date, Pertinent Pages, Etc.)

	"SERIE ISOMORPHE D'ORTHOSILICATES ( $T_2SiO_5$ ) ET D'ORTHOGERMANATES ( $T_2GeO_5$ ) DE TERRES RARES", Buisson et al., Mat. Res. Bull. Vol. 1 pp. 193-198, 1968.
	"PREPARATION AND CATHODOLUMINESCENCE OF $Ca^{3+}$ -ACTIVATED YTTRIUM SILICATES AND SOME ISOSTRUCTURAL COMPOUNDS", A.H. Gomes de Mesquita et al., Mat. Res. Bull. Vol. 4, pp. 643-650, 1969.
	"GROWTH OF LANTHANIDE OXYORTHOSILICATE SINGLE CRYSTALS, AND THEIR STRUCTURAL AND OPTICAL CHARACTERISTICS", G.V. Anan'eva et al., Translated from Izvestiya Akademii Nauk SSSR, Neorganicheskie Materialy, Vol. 17, No. 6, pp. 1037-1042, June, 1981.

Examiner

Date Considered

\*Examiner: Initial citation considered, whether or not citation is in conformance with HPEP 609; Draw line through citation if not in conformance and not considered. Include copy of this form with next communication to applicant.

SMS003270



Sheet 2 of 4

Form PTO-1449 (REV. 2-82)	U.S. Department of Commerce Patent and Trademark Office	Atty. Docket No. 27515-539/11809	Serial No. 07/254,351
INFORMATION DISCLOSURE STATEMENT BY APPLICANT (Use several sheets if necessary)		Applicant Charles L. Melcher	
		Filing Date October 6, 1988	Group 113

[illegible][illegible]

OTHER DOCUMENTS (including Author, Title Date, Pertinent Pages, Etc.)	
	"SITE SELECTIVELY EXCITED LUMINESCENCE OF $\text{Eu}^{3+}$ IN GADOLINIUM, YTTRIUM AND LUTETIUM OXYORTHOSILICATES", Halse et al., Journal of the Less-Common Metals, 126 (1986) 215-220.
	"CERIUM-ACTIVATED $\text{Gd}_2\text{SiO}_5$ SINGLE CRYSTAL SCINTILLATOR", Takagi et al., Appl. Phys. Lett., Vol. 42, No. 1, 1 January 1983, pp. 43-45.
	"CZOCHERALSKI GROWTH OF RARE-EARTH ORTHOSILICATES ( $\text{Ln}_2\text{SiO}_5$ )", Brandle et al., Journal of Crystal Growth 79 (1986) pp. 308-315.

Examiner	Date Considered
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\*Examiner: Initial citation considered, whether or not citation is in conformance with NPEP 609; Draw line through citation if not in conformance and not considered. Include copy of this form with next communication to applicant.







Sheet 4 of 4

Form PTO-1449  
(REV. 2-82)

U.S. Department of Commerce  
Patent and Trademark Office

Atty. Docket No. 27515-539/11889	Serial No. 07/254,353
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Serial No.  
07/254 153

INFORMATION DISCLOSURE STATEMENT  
BY APPLICANT  
(Use several sheets if necessary)

Applicant  
Charles L. Melcher

Filing Date October 6, 1988	Group 113
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## U.S. PATENT DOCUMENTS

[illegible]

FOREIGN PATENT DOCUMENTS

[illegible]

OTHER DOCUMENTS (including Author, Title Date, Pertinent Pages, Etc.)

[illegible]

**Examiner**

Date Considered	Number of Cases	Number of Deaths	Number of Recoveries	Number of Discharges	Number of Deaths	Number of Recoveries	Number of Discharges
1918	100	10	80	10	10	80	10
1919	100	10	80	10	10	80	10
1920	100	10	80	10	10	80	10
1921	100	10	80	10	10	80	10
1922	100	10	80	10	10	80	10
1923	100	10	80	10	10	80	10
1924	100	10	80	10	10	80	10
1925	100	10	80	10	10	80	10
1926	100	10	80	10	10	80	10
1927	100	10	80	10	10	80	10
1928	100	10	80	10	10	80	10
1929	100	10	80	10	10	80	10
1930	100	10	80	10	10	80	10
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1969	100	10	80	10	10	80	10
1970	100	10	80	10	10	80	10
1971	100	10	80	10	10	80	10

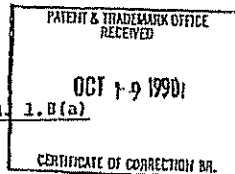
\*Examiner: Initial citation considered, whether or not citation is in conformance with MPEP 609; Draw line through citation if not in conformance and not considered. Include copy of this form with next communication to applicant.

SMS003273



THE UNITED STATES PATENT AND TRADEMARK OFFICE

PATENT  
A27515-1  
539/15132



CERTIFICATE OF MAILING UNDER 37 C.F.R. 1.8(a)

Paper Certificate of Correction

Patent  
Serial No. 4,958,080  
Issue  
Filing Date Sept. 18, 1990

I hereby certify that this paper is being deposited  
with the United States Postal Service as first class mail in  
an envelope addressed to:

Commissioner of Patents and Trademarks  
Washington, D.C. 20231

on October 1, 1990  
Date of Deposit

October 1, 1990  
Date of Signature

Arthur S. Tenser  
Attorney Signature

Arthur S. Tenser  
Attorney Name

18,839  
Registration No.

SMS003274

Printers Only  
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UNITED STATES PATENT AND TRADEMARK OFFICE  
CERTIFICATE OF CORRECTION

PATENT NO. : 4,950,050

DATED : September 13, 1990

INVENTOR(S) : CHARLES L. NELCHER

It is certified that error appears in the above-identified patent and that said Letters Patent is hereby corrected as shown below:

First page, Item 63, AND col: 1, line 6, after "continuation"  
insert -- in part --  
col. 1, line 6, delete "now abandoned",  
col. 1, line 7, after "1950" insert -- , now abandoned --

SMS003275

UNITED STATES PATENT AND TRADEMARK OFFICE  
CERTIFICATE OF CORRECTION

PATENT NO. : 4,956,080

DATED : September 18, 1990

INVENTOR(S) : Charles L. Helcher

It is certified that error appears in the above-identified patent and that said Letters Patent is hereby corrected as shown below:

Title page, item (63), AND col. 1, line 6, after "continuation"  
insert -- in part --;

Col. 1, line 6, delete "now abandoned":

Col. 1, line 7, after "1988" insert -- , now abandoned --.

Signed and Sealed this  
Ninth Day of June, 1993

Attest:

DOUGLAS B. COMER

Acting Officer

Acting Commissioner of Patents and Trademarks

SMS003276

BRUMBAUGH, GRAVES, DONOHUE & RAYMOND

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NEW YORK, N.Y. 10118

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October 17 1990

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MICHAEL S. BOWEN  
MICHAEL S. BOWEN  
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MICHAEL S. BOWEN  
MICHAEL S. BOWEN  
MICHAEL S. BOWEN  
MICHAEL S. BOWEN  
MICHAEL S. BOWEN

ANDREW H. KENNEDY  
LOUIS S. KENNEL  
STANLEY S. FORDICE III  
ROBERT W. HANCOCK

380 MANHATTAN HOTEL PARKWAY  
HAUPPAUGE, LONG ISLAND, N.Y. 11787  
(516) 430-4411

CORRESPONDENCE  
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WRITER'S DIRECT DIAL NUMBER

212 400-2542

Honorable Commissioner of Patents  
and Trademarks  
Washington, D.C. 20231

Re: U.S. Patent No. 4,958,080  
Issued: September 18, 1990  
"LUTETIUM ORTHOSILICATE SINGLE  
CRYSTAL SCINTILLATOR DETECTOR"  
Inventor: Charles L. Melcher  
(Our File A27515-1 939/15132)

Sir :

Upon comparison of Patent No. 4,958,080 granted September 18, 1990 to Charles L. Melcher for "Lutetium Orthosilicate Single Crystal Scintillator Detector", with our file of the application therefor, errors in printing were found. Enclosed herewith is a proposed Certificate of Correction in duplicate.

The correct forms of the errors made in printing appear at the following places in the application:

Examiner's Amendment dated February 26, 1990  
page 3.

It is respectfully requested that the Certificate of Correction be issued for attachment to the original patent under the provisions of 37 CFR 1.322.

Respectfully submitted,

*Arthur S. Tenser*  
Arthur S. Tenser  
Patent Office Reg. No. 18,839

Enclosure

SMS003277

## NOTICE RE: CERTIFICATES OF CORRECTION

Page No. 15

DATE : 4/15/92  
 TO : Supervisor, Art Unit *4 HIB H.H.*  
 SUBJECT : Certificate of Correction Request in Patent No. *4,958,080*

A response to the following question(s) is requested with respect to the accompanying request for a certificate of correction.

- ☐ 1. Would the change(s) requested under 37 CFR 1.323 constitute new matter or require reexamination of the application?
- ☐ 2. Would the change(s) requested under 37 CFR 1.323 materially affect the scope or meaning of the claims allowed by the examiner in the patent?
- ☐ 3. Applicant disagrees with change(s) initiated and dated by Examiner in lieu of an Examiner's Amendment. Should the change request be granted?
- ☒ 4. *Is this a continuation in part.* With respect to the change(s) requested, correcting Office errors, should the patent read as shown in the certificate of correction?
- ☐ 5. If the amendment filed \_\_\_\_\_ had been considered by the Examiner, would the amendment have been entered?

PLEASE RESPOND WITHIN 7 DAYS AND RETURN THE FILE TO  
 ROOM 809, PKJ

*Sean Seng*  
 Patent Assistant

## TO: CERTIFICATES OF CORRECTION BRANCH

DATE: 4/30/92

The decision regarding the change(s) requested in the certificate of correction is shown below.

- |  |                             |   |
|--|-----------------------------|---|
| 1. <input checked="" type="checkbox"/> YES | <input type="checkbox"/> NO | <input type="checkbox"/> Comments below |
| 2. <input type="checkbox"/> YES            | <input type="checkbox"/> NO | <input type="checkbox"/> Comments below |
| 3. <input type="checkbox"/> YES            | <input type="checkbox"/> NO | <input type="checkbox"/> Comments below |
| 4. <input type="checkbox"/> YES            | <input type="checkbox"/> NO | <input type="checkbox"/> Comments below |
| 5. <input type="checkbox"/> YES            | <input type="checkbox"/> NO | <input type="checkbox"/> Comments below |

☐ Comments \_\_\_\_\_

\_\_\_\_\_  
 \_\_\_\_\_  
 \_\_\_\_\_

*Patricia M. [Signature]*  
 Supervisor

*H.H.*  
 Art Unit

PTOL-300 (REV. 10/87)

U.S. DEPARTMENT OF COMMERCE Patent and Trademark Office

SMS003278

**EXHIBIT 8**



# Boron Nitride, A Neutron Scintillator With Deficiencies

R. Engels, Member, IEEE, G. Kemmerling and J. Scheiten

**Abstract**—During an electron beam evaporation accidentally sintered boron nitride was discovered to be a bright scintillating material [1]. The high density boron nitride (HDBN) of Henze [2] showed most promising results concerning light emission due to 8 keV electron radiation, due to bombardment with 5 MeV alpha particles, and due to 240 nm UV radiation. The light pulses generated in HDBN are similar in intensity as those generated in the well known  $^6\text{Li}$  glass scintillator. Because of the extremely strong absorption of thermal neutrons and the weak interaction with MeV gammas the material is an excellent new inorganic scintillator candidate for the detection of thermal and epithermal neutrons. A series of properties of HDBN was measured with various methods and evaluated. Many results are in favour of an intended use of HDBN as scintillator. A few observations reveal deficiencies of this new scintillator and ask for further investigations.

## 1. INTRODUCTION

A material which heavily absorbs thermal neutrons and which emits a bright light pulse due to such an absorption event can be utilized as scintillator for the detection of neutrons. The scintillators are optically coupled with photomultipliers or semiconductor diodes which then generate an electrical pulse for each absorbed neutron. For a reliable, position sensitive detection with high spatial resolution a series of properties must exist. They are listed in the following:

### A. Absorption probability

A well designed scintillator should stop thermal neutrons with a probability of 70%. At this level the ratio of signals generated by thermal neutrons to noise generated by fast neutrons is optimised.

### B. Colour of scintillation light

For blue light with a wave length of about 400 nm the light guidance as well as the generation of photo electrons and of ion pairs in photomultipliers and photodiodes, respectively, are performed most efficiently.

### C. Number of photons per absorbed neutron

If a neutron is absorbed in  $^6\text{Li}$ ,  $^{10}\text{B}$ , and  $^3\text{He}$  nuclei an energy of 4.78 MeV, 2.31 MeV, and 0.78 MeV is released, respectively. Only a small fraction of typically less than 1% is converted into light energy. The vast majority of energy dissipates as heat. At least 1000 photons per neutron should

arrive at the photo multiplier or photo diode in order that a well defined event position can be deduced.

### D. Duration of light pulses

The decay time of a generated light pulse should be not more than 100 ns in order that pile-up effects can be ignored even for pulses which rapidly follow each other. In addition, such short pulses do not affect the temporal time structure as determined by choppers and pulsed sources.

### E. Optical transparency

For optically transparent scintillators one can expect sharp light pulses. In non-transparent scintillators the pulse height spectra become broad due to multiple reflection and self absorption. With very broad spectra neutron pulses occur which fall beyond a lower level discrimination threshold and are not counted.

### F. Gamma insensitivity

In scattering experiments with thermal neutrons a strong flux of gamma quanta with energies in the MeV region cannot be avoided. This flux is generated in the neutron source, which might be a fission reactor or a spallation source; it is also generated when the primary beam is monochromatized, pulsed, collimated, guided and scattered in the sample. A residual sensitivity of 1 ppm seems to be tolerable, a value of 100 ppm is not acceptable as was experimentally verified quite often. Usually low gamma sensitivity of scintillators is achieved by pulse height discrimination. Attempts to obtain additional gamma discrimination by pulse shape analysis were successful in special cases with a huge amount of electronic equipment.

### G. Light background

A continuous weak after-glow in the scintillator material leads to a strong wing at low pulse height in the pulse height spectrum. There, the pulse rate decreases exponentially and rapidly towards zero with increasing pulse height. Usually a discrimination threshold against gamma rays is set so high that it completely repels these signals due to this light background.

### H. Large detector areas

Scattering experiments can profit from the circumstance that the sources suffer from weak neutron fluxes but offer big source areas. The profit can be orders of magnitude in scattered intensity provided large area detectors are available. Large means that the detector cross sections are of the order of 100 to 10000 cm<sup>2</sup>. So far the largest scintillation detectors are not more than 2500 cm<sup>2</sup>.

Manuscript received November 7, 2005.

R. Engels, and G. Kemmerling with the Zentralinstitut für Elektronik; Forschungszentrum Jülich GmbH, 52425 Jülich, Germany (email: r.engels@fz-juelich.de; g.kemmerling@fz-juelich.de).

J. Scheiten with the Institut für Schichten und Grenzflächen; Forschungszentrum Jülich GmbH, 52425 Jülich, Germany (email: j.scheiten@fz-juelich.de).

### 1. Scintillator cost

A discussion of the cost-to-benefit relation for neutron detectors asks for large area detectors. In view of this, a scintillator of a low specific price (cost-per-unit area) is extremely welcome.

A scintillator material which simultaneously meets all these nine requirements does not exist. The following brief discussion demonstrates, that only very few scintillator types are useful for neutron detection.

#### 1) Li glass

This is essentially the only scintillator which has been used in large area scintillation detectors for thermal neutrons. The neutron absorption takes place in  $^6\text{Li}$  nuclei. By blending  $^6\text{Li}$ -enriched  $\text{Li}_2\text{O}$  into the glass typical to an amount of 18% by weight about 1mm thick scintillator plates are able to stop thermal neutrons with the required probability of 70%. The product is commercially available from Applied Scintillation Technologies [3].

#### 2) LiGdBorate

A brighter neutron scintillator was developed by Photogenics, Salt Lake City, Utah, USA. The material is  $^6\text{Li}^{13}\text{Gd}^{11}\text{Borate}$ . It is the role of the isotope  $^6\text{Li}$  to absorb thermal neutrons, while the isotopes  $^{13}\text{Gd}$  and  $^{11}\text{B}$  are chosen to prevent neutron absorption by Gadolinium and Boron. The crystalline borate powder is bedded into an epoxy resin which has the same refractive index as the borate [4]. Due to such a match an optical transparency should be achieved. In an ultimate step, the epoxy resin must be deuterated in order to avoid the strong incoherent neutron scattering of hydrogenous materials. The development of this scintillator was subsidized for more than ten years. Meanwhile the financial support has stopped [5]. So far the scintillator was used once in a large area scintillation detector at ISIS because of its intense light response on a neutron absorption event.

#### 3) Li-Iodide

A scintillator with excellent physical properties is single crystalline  $^6\text{Li}$ -iodide. A few single crystal disks of 4cm diameter which are encapsulated by glass do world wide exist.

For a long time no further LiI single crystals were grown and no new  $^6\text{Li}$  neutron scintillators were on the market.

#### 4) BCS23A

This liquid scintillator is commercially available from Bicorn, now Saint Gobin, Crystals and Detectors [6]. The neutrons are absorbed by the  $^{10}\text{B}$ -enriched boron component of the organic liquid. The liquid is optically transparent and chemically instable. Its light output is weak. Again, the hydrogenous organic molecules must be deuterated in order to get rid of the strong incoherent neutron scattering.

#### 5) Other scintillators

There are more organic liquids and small single crystals which absorb neutrons to some extent and scintillate upon ionising radiation. In some respect they have fantastic properties, however, they exist only as laboratory samples and are used for scintillator studies only.

More information is given by C.W.E. van Eijk [7].

In view of the nine requirements which are described in the first part of this chapter all five scintillators listed above have more or less severe deficiency.

The main disadvantage of the Li glass (1) scintillator is its insufficient gamma insensitivity which is in the order of 100 ppm for MeV gammas. One would like to have more intense light pulses in order that there are more than the measured 2000 photons per neutron absorption event at the photo cathode which directly influences the spatial resolution in Anger type detectors [8]. In addition, the scintillator cost with 3000 EUR / 100 cm<sup>2</sup> is large.

For the  $^6\text{Li}^{13}\text{Gd}^{11}\text{Borate}$  scintillators the deficiencies are more severe. The poor transparency causes broad pulse height spectra in order that a pulse height discrimination against gamma quanta and electronic noise affects already the detection of neutrons. The used Gd isotopes  $^{13}\text{Gd}$  and  $^{160}\text{Gd}$  are extremely expensive and they are available in only small quantities. A homogenisation of the powder/resin mixture was not successful on microscopic scale. Financial support of further developments had come to an end and a commercial use is not foreseen.

The powerful semiconductor industry has lead to single crystalline silicon waver with 300mm diameter. A similar development is not expected for lithium iodide. It is more likely that the growth of  $^6\text{Li}$ I single crystal has stopped for ever and that the development of neutron scintillation detectors must look after alternatives of  $^6\text{Li}$ I scintillators.

With liquid scintillators the light signals are too small, the pulse height spectra are too broad and the differences in pulse heights between neutron and gamma signals are too little.

In this context a new inorganic scintillator is introduced. The scintillation property of high density boron nitride (HDBN) which is fabricated by sintering BN powder at high isostatic pressure and elevated temperature was discovered fortuiously by Angelika Prucht while she was making gold pads on HDBN plates by electron beam evaporation. The gold pads were used for transport measurements in HDBN. A day later a piece of Li glass scintillator was mounted next to a HDBN sample in the electron beam evaporator. With the electron beam switched on but the shutter still closed both samples emitted light visible with the bare eyes. The HDBN piece appeared brighter than the well known  $^6\text{Li}$  glass scintillator. The scintillation in both samples was probably caused by X ray Bremsstrahlung which is generated in the electron beam heater and which circumvented the shutter above the evaporation source. The observation stimulated a lot of experiments in order to get to an evaluation HDBN as neutron scintillator.

It is the purpose of this paper to describe these experiments and to evaluate the results. This ends with a list of favourable properties of HDBN, with some recognized deficiencies for using HDBN as neutron scintillator and a few suggestions how to proceed.

It is hoped that the presentation of these preliminary results will lead to some stimulating discussions and fruitful cooperation.

## II EXPERIMENTAL RESULTS

In a Scanning Electron Microscope SEM the cathode luminescence detector was used to measure the spectral distribution of the emitted light as generated by the radiation with 8 keV electrons in HDBN and Li glass. In both cases well pronounced intensity peaks accrued at 400 nm with a FWHM of  $\sim 100$  nm due to this near-surface excitation. The peak heights of both scintillators were also comparable.

In the next experiment the two materials were radiated by 5 MeV alphas from a radioactive  $^{241}\text{Am}$  source in front of a photo multiplier. The near-surface excitation generates light pulses which are half the size in HDBN compared to Li glass. The pulse height distribution is peaked in both scintillators, however in the transparent Li glass the distribution is much sharper. Fig. 1 shows the alpha peak as measured with the HDBN.

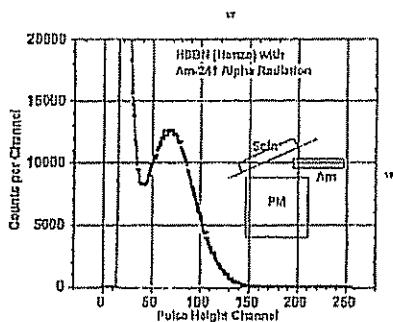


Fig. 1 Pulse height spectrum caused by alpha radiation of HDBN in the indicated  $45^\circ$  geometry

Because of the non-transparent HDBN the  $45^\circ$  geometry was chosen. The strong wing at low pulse height is an after-glow effect.

Day light exposure leads to the strong after-glow which is visible by eye many hours later in the dark. The decay of this after-glow occurs with different rates over hours, days, and even over weeks.

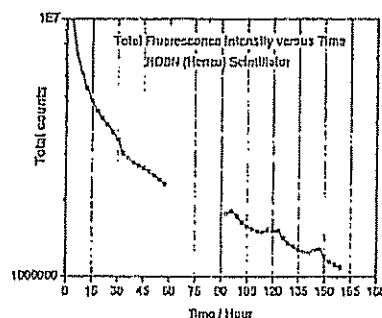


Fig. 2 Long time decay of after glow in HDBN

Fig. 2 shows the long time behaviour as measured with a photo multiplier.

The temporal width of a light pulse due to an alpha trace is very short, a half width of the temporal peak is only 2.5 ns as indicated by the oscilloscope diagram in Fig. 3.

An annealing of HDBN at 500°C for 1 hour destroys the after-glow completely.

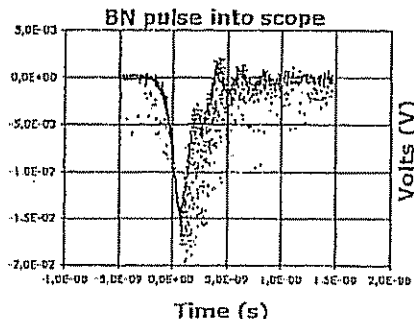


Fig. 3 Temporal pulse shape as measured with a digital storage oscilloscope

Further experiments were done with frequency-doubled laser light of 5.1 eV energy and 240 nm wave length. The source power was 17 mW. The main results are:

1. HDBN fluoresces orders of magnitude stronger than BN powder which is the starting material of HDBN.
2. HDBN fluoresces similarly strong as Li glass or ordinary quartz glass.
3. Part of the light generated in HDBN decays very slowly within hours or even days. Such behaviour was not observed in other substances.

Some properties of boron nitride are compiled in Table 1. An extremely favourable property of HDBN is its efficient neutron absorption. HDBN with  $^{10}\text{B}$ -enriched boron has an absorption length of 50  $\mu\text{m}$ . The ranges of the two nuclei, which are released when a neutron is absorbed, are 3.9  $\mu\text{m}$  and 2.15  $\mu\text{m}$ . HDBN is a low Z material with 12 electrons per unit cell containing a boron and nitrogen atom. The scintillation light has an energy of  $\sim 3$  eV which is smaller than the gap energy. Thus there is no self-absorption in pure BN. Pure crystalline BN has a gap energy of  $\sim 7.5$  eV [9] and is probably an indirect semiconductor.

From a full chemical analysis no components were found which could play the role of activation centres. Contaminants were found only in the ppm range. In other inorganic crystalline scintillators such activation centres are deliberately created with additives grown into the crystals. In the case of HDBN it is more likely that activation centres are formed by lattice defects in the sintering zone near the surface of the small crystalline BN grains. There the lattice periodicity is gradually distorted which leads to a reduction of gap energy. Without a drastic reduction of the gap energy one would not reach excited states by 240 nm UV radiation. In addition the defects form localized lattice states where the transition from an excited state to its ground state can occur more likely via the emission of light.

TABLE I  
DATA OF BORON AND BORON NITRIDE

Boron Isotope		$^{10}\text{B}$	$^{11}\text{B}$
Abundance		20%	80%
Absorption cross section for 2200 m/sec (thermal) neutrons	$\sigma$	3835 barn	0.0055 barn
Molecular weight of Boron	$M_B$	10.80	
Molecular weight of BN	$M_{BN}$	24.80	
HDBN mass density	$\rho$	2.10 g/cm <sup>3</sup>	
Atomic number density	$n$	$0.051 \cdot 10^{24} \text{ cm}^{-3}$	
Absorption coefficient	$\mu$	$195.5 \text{ cm}^{-1}$	
Scintillator thickness for natural boron nitride	$d$	255 $\mu\text{m}$	
Scintillator thickness for $^{10}\text{B}$ N	$d^*$	51 $\mu\text{m}$	
Neutron reaction to 94%		$^{10}_0\text{n} + ^{10}_5\text{B} \rightarrow ^4_2\text{He} + ^7_3\text{Li}^* + 2.31 \text{ MeV}$	
Energy distribution		$E(\text{He}) = 1.47 \text{ MeV}$	$E(\text{Li}) = 0.84 \text{ MeV}$
Range of reaction nuclei in boron nitride		$R(^4\text{He}) = 3.9 \mu\text{m}$	$R(^7\text{Li}) = 2.15 \mu\text{m}$

### III SUMMARY AND CONCLUSIONS

In the introduction nine requirements are discussed which should be met by an ideal neutron scintillator. In view of these the HDBN solid is evaluated

#### 1) Absorption Probability

$\text{HD}^{10}\text{BN}$  is a very strong neutron absorber. The absorption length is only 50  $\mu\text{m}$ .

#### 2) Colour of scintillation light

The mean light wave length is 400nm, ideal for photomultipliers and semiconductor diodes

#### 3) Number of photons per absorbed neutron

A preliminary estimate leads to 6 to 8 time less intense signals compared to  $^6\text{Li}$  glass. This, however, is not yet based on a direct measurement. So far no attempts are being made to increase the light output

#### 4) Duration of light pulses

The temporal width is extremely small, only 2.5 ns which is sufficient for any time of flight measurement and is favourable in avoiding pile-up effects

#### 5) Optical transparency

Imm thick HDBN appear white like salt in a bag. Day light is reflected in HDBN at the sintered BN grains like it is reflected in salt bag at the piled-up salt grains. In both cases there is no light absorption

#### 6) Gamma Insensitivity

There are sound arguments that the gamma sensitivity is very low. BN is low Z material and the required scintillator thickness is extremely small. Thus the probability of a photo effect, Compton process or pair production is small; in addition, the created high energy electron can hardly deposit its energy in the thin scintillator. The gamma sensitivity has not been measured yet

#### 7) Light background

The after-glow is very strong, however, one gets rid of this light emission after annealing the scintillator at 500C for 1h. This procedure might be inconvenient, however, it is very successful

#### 8) Large detector areas

HDBN is available in large quantities of different shape and size. HDBN can be shaped by cutting, milling, lacing, polishing, and drilling.

#### 9) Scintillator cost

The material HDBN is cheap compared to any other scintillator

At present it is too early to give more detailed information on this topic

The deficiencies of this scintillator material are:

The low light output per absorbed neutron which is serious drawback

- 1) The strong after glow which disappears after annealing
- 2) The optical non transparency based on reflection but probably not on absorption at grains.
- 3) For the next experiments which are under preparation HDBN disks of 2mm thickness are thinned down to 0.05 mm

With such thin scintillators we should be able to measure directly the pulse height and pulse height distributions of light pulses generated by neutron absorption. Before exposure to a neutron beam the thin HDBN disks will be annealed in a light tight container and mounted in front of a photo cathode of a photo multiplier under IR illumination.

In an other experiment the transparency of the thin HDBN disks will be investigated in order to show that there is no absorption but solely reflection

In further experiments the light response is investigated by high temperature treatments where atomic diffusion can take place

### ACKNOWLEDGEMENT

We are thankful to H. P. Boehm for cathode luminescence measurements, to Dr. G. Creeljus for the UV radiation experiments and valuable discussions, to Dr. David E. Holcomb, ORNL for performing two interesting experiments with HDBN and for his interest in this subject

### REFERENCES

- (1) Angelika Pracht, private communication about her discovery of the scintillation of HDBN, Mar 2001

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- [3] 'Li glass scintillators are manufactured by Applied Scintillation Technologies[3], Harlow ( UK ). For more details see <http://www.space-science.com/esit/>
- [4] J.B. Cairr, G.M. MacGillivray, R.R. MacGillivray, P.J. Seddon. "Performance and characteristics of a new scintillator", Nucl. Instr. Meth., A 424 (1999), 15-19
- [5] Dave Merrill Photogenics, Salt Lake City, Utah ( USA ) private communication (2004)
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- [7] C.W.E.van Eljk , Review on "Inorganic Scintillators and Storage Phosphors for Position Sensitive Neutron Detection" in the Conference Proceeding of the workshop on Position Sensitive Neutron Detectors, Berlin, Germany, June 28-30 (2001)
- [8] R. Engelr, E. Janzen, R. Reinartz, P. Reihardt, W. Schöfer, and J. Scheibel IEEE Transactions on Nuclear Science, 45 (1999) 502-504 "Realization of a Small-Size High Resolution Linear Scintillation Detector"
- [9] See, S.M., Physics of Semiconductor Device, New York, Wiley Interscience Publication, 1981, pp 848-849

**EXHIBIT 9**

From:  
Sent: Wednesday, March 8, 2006 9:29 AM  
To:  
Subject: FW: Philips GEMINI TF

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REDACTED

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From: Rothan, Dominique  
Sent: Tuesday, March 07, 2006 5:20 PM  
To:   
Subject: Philips GEMINI TF

It's out there! Interesting to see their price premium and their volume projection for 06.

**Philips launches new PET/CT product, ultrasound upgrades at ECR**  
3/6/2006

VIENNA - Multimodality developer Philips Medical Systems of Andover, MA, unveiled a new PET/CT and a new ultrasound system at the European Congress of Radiology (ECR) this week.

The Gemini TF PET/CT system uses gamma ray time measurements to deliver an increase in image quality and consistency, according to the firm. The device is said to raise effective image sensitivity by more than two times over conventional PET, and can conduct image acquisition of patients of any body size in less than 10 minutes for a whole-body PET scan, the company said.

The device utilizes a lutetium yttrium orthosilicate (LYSO) scintillator crystal, curved photomultipliers, and Philips' proprietary TruFlight technology, which uses the time difference -- in pico seconds ( $10^{-12}$  seconds) -- between the detection of coincident events to more accurately identify the origin of the annihilation.

Consistent with all Gemini PET/CT systems, the Gemini TF also features Philips' OpenView gantry design, which allows for easier administration of radiopharmaceuticals for myocardial perfusion studies, patient monitoring equipment leads, and patient comfort, according to the company.

The system received its 510(k) clearance from the Food and Drug Administration in November 2005, and is scheduled for release in the second quarter this year. Company representatives said that Gemini TF will be available in 16- and 64-slice CT models and will be priced from \$2.7 million to \$3.4 million, depending on configuration.

Philips expects to sell approximately 40 Gemini TF systems worldwide in 2006, representatives said.

The firm also demonstrated its IU22 ultrasound system, which features new upgrades and enhancements including improved cardiology capabilities and detailed 3D and 4D fetal imaging. The IU22 utilizes the firm's new L9-3 transducer with xMatrix technology and its QLab quantification software.

Ergonomic factors have been incorporated into all areas of the product's design, allowing for a fully pivoting and extendable viewing screen, as well as voice controls that accept 2,200 individual commands in five languages: English, German, Spanish, French, and

SGCP006210

Italian. The voice capabilities take approximately five minutes of training to accept a new user's voice pattern, according to company representatives.

Dominique

REDACTED

<http://www.detecteurs.saint-gobain.com/>

SGCP006211



EXHIBIT 10  
REDACTED  
IN ITS ENTIRETY

**EXHIBIT 11**

**SIEMENS**

via Telefax +31 40 27 43489

Philips Intellectual Property & Standards  
Attn: Mr. Marc Schouten  
P.O. Box 220  
5600 AE Eindhoven

NETHERLANDS

Name  
Abteilung  
Standort  
Telefon  
Fax  
E-Mail

Heinz Schmidt  
CT IP Med  
ERL S SC

schmidt.heinz@siemens.com

Ihr Schreiben  
Unser Zeichen

Sid / BMD  
17 March 2006

Datum

**REDACTED**

Dear Mr. Schouten,

As you may know, Siemens is the exclusive licensee of United States Patent No. 4,958,000 by virtue of its acquisition of CTI Molecular Imaging, Inc. last year. This patent relates to a gamma ray or x-ray detector having a scintillator composed of the material lutetium oxyorthosilicate ("LSO"). We enclose a copy of the relevant patent for your convenience.

We have recently learned that your "Gemini Raptor" PET/CT system uses a detector scintillator composed of lutetium yttrium oxyorthosilicate ("LYSO") that we believe infringes this patent. This is disconcerting as this patent is deemed excluded under our existing patent cross-license agreement. Siemens has closely investigated LYSO as a scintillator material. We believe there is no substantive difference between the two materials designated LSO and LYSO. The investigation of Siemens regarding the properties of LYSO is born out by a recently published paper, entitled "Large Size LYSO Crystals for Future High Energy Physics Experiments," IEEE Trans. Nucl. Sci. NS-52 (2005) 3133. This paper states in its conclusion "Ce doped LSO and LYSO crystals have identical emission, excitation and transmission spectra."

Further, Siemens is prosecuting a litigation concerning the European equivalent of the United States patent, European Patent No. 0 373 976, against Photonic Materials Ltd. in the Court of Sessions of Edinburgh, Scotland for making and selling LYSO material. Siemens has every expectation that this litigation will be resolved shortly in its favor.

**Corporate Technology**

Corporate Intellectual Property and Functions

Leitung:  
Dr. Winfried Böllner

Briefadresse:  
Siemens AG

Postfach 22 16 34  
80506 München

Hausadresse:  
San-Carlos-Str. 7  
81058 Erlangen

Siemens Aktiengesellschaft, Vorsitzender des Aufsichtsrats: Heinrich v. Pierer, Vorstände: Klaus Klotzfeld, Vorsitzender, Johannes Feldmayer, Thomas Gernreich, Edward G. Kuebeck, Rudi Lamprecht, Hans-Joachim Hübner, Jürgen Rademacher, Eberhard R. Reikhardt, Udo J. Storz, Claus Weylich, Hans-Wilhelm Storz, Sitz der Gesellschaft: Berlin und München, Registergericht: Berlin-Charlottenburg, HRB 122807, München, HRB 16264

PH 1169

IDNR: 4210/Y: 99-1.00/BVai

SIEMENS

REDACTED

Page 2 to letter of 17 March 2006  
to Philips Intellectual Property

we  
are very concerned about the introduction of the LYSO scanner by Philips. We believe it would  
be advantageous to meet with representatives of Philips to discuss this matter so that we may  
try to resolve this issue. Siemens feels strongly that such a meeting is necessary before Philips  
makes plans for broad commercial marketing for such a system which potentially infringes our  
above-referenced patent.

We look forward to hearing from you at your earliest convenience.

Siemens Aktiengesellschaft

  
Schmidt

sgd. ppa Kirschbaum

Enc.

US 4,958,080

PH 1170

**EXHIBIT 12**

## Large Size LYSO Crystals for Future High Energy Physics Experiments

Hanning Chen, Liyuan Zhang Member, IEEE and Ren-yuan Zhu Senior Member, IEEE

**Abstract**—Because of high stopping power and fast bright scintillation, cerium doped silicate based heavy crystal scintillators, such as GSO, LSO and LYSO, have been developed for medical instruments. Their application in high energy and nuclear physics, however, is limited by lacking high quality crystals in large size. The optical and scintillation properties, including the transmittance, emission and excitation spectra and the light output, decay kinetics and light response uniformity as well as their degradation under irradiation, were measured for large size LYSO samples from different vendors, and were compared to a BGO sample of the same size. Possible applications for crystal calorimetry in future high energy and nuclear physics experiments are discussed.

**Index Terms**—Lutetium Oxyorthosilicate; Lutetium Yttrium Oxyorthosilicate; Crystal; Scintillator; Transmittance; Emission; Light Output; Radiation Damage.

### I. INTRODUCTION

IN the last decade, cerium doped silicate based heavy crystal scintillators have been developed for the medical industry. As of today, mass production capabilities of Gadolinium Oxorthosilicate ( $Gd_2SiO_5$ , GSO) [1], Lutetium oxyorthosilicate ( $Lu_2SiO_5$ , LSO) [2] and Lutetium yttrium oxyorthosilicate ( $Lu_{1-x}Y_xSiO_5$ , LYSO) [3], [4] were established. Table I lists basic properties of commonly used heavy crystal scintillators; NaI(Tl), CsI(Tl), undoped CsI, BaF<sub>2</sub>, bismuth germanate ( $Bi_4Ge_3O_{12}$  or BGO) and lead tungstate ( $PbWO_4$  or PWO) as well as cerium doped GSO and LSO. All these crystal scintillators, except LSO and GSO, have been used in high energy or nuclear physics experiments. An early example is a Crystal Ball NaI(Tl) calorimeter at SPEAR. CsI(Tl) crystals were used for calorimeters of CLEO at CESR, BABAR at SLAC and BELLE KEK. KTeV built a CsI calorimeter at Tevatron, and L3 built a BGO calorimeter at LEP. BaF<sub>2</sub> crystals were used in the TAPS experiment at GSI, and were proposed by the L<sup>\*</sup> and GEM experiment at SSC. Recently, PWO crystals are used by CMS and ATLAS at LHC, by BTeV at Tevatron and by CLAS and PrimEx at CEBAF.

Because of high stopping power and fast bright scintillation light, cerium doped silicate has also attracted a broad interest in the physics community. The main obstacles of using these crystals in experimental physics are two fold: the availability of high quality crystals in adequate size and the high cost

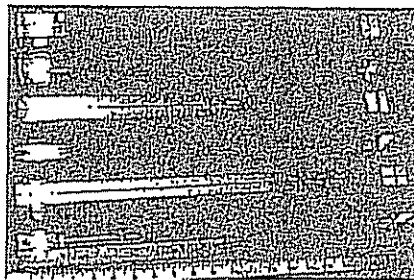


Fig. 1. A photo showing three large size (2.5x2.5x20 cm) scintillation crystal samples. From top to bottom: SIC BGO, CPI LYSO and Saint-Gobain LYSO.

associated with their high melting point ( $\sim 2000^\circ\text{C}$ ). Recent emergence of large size LYSO crystals in market, however, inspired this investigation on possible application of this new generation scintillators in experimental physics, such as a super B factory [5].

Fig. 1 is a photo showing three large size (2.5x2.5x20 cm) crystal samples. They are, from top to bottom: a BGO sample from Shanghai Institute of Ceramics (SIC) and two LYSO samples from Crystal Photonics, Inc. (CPI) and Saint-Gobain Ceramics & Plastics, Inc. (Saint-Gobain). While the SIC and Saint-Gobain samples have perfect geometry and surface polishing, the CPI sample has chips at corner and surface as shown in Fig. 1. This is due to the fact that CPI does not have adequate polishing and treatment facilities for such large size samples. To facilitate a comparison of basic optical and scintillation properties, samples of 1.5 radiation length (1.7 cm) cubic were also measured: two BGO samples from SIC, two LSO samples from CIT and two LYSO samples each from CPI and Saint-Gobain.

All surfaces of these samples are polished. No thermal treatment was applied before measurement. The transmittance spectrum was measured by using a Hitachi U-3210 UV/visible spectrophotometer with double beam, double monochromator and a large sample compartment equipped with a custom Helon coated integrating sphere. The systematic uncertainty in repeated measurements is about 0.3%. Taking into account multiple bouncing between two end surfaces, the theoretical limit of transmittance without internal absorption,  $T_{\infty}$ , can be

This work was supported in part by the U.S. Department of Energy Grant No. DE-FG01-93-ER40701.

Ren-yuan Zhu is with the California Institute of Technology. Tel.: (626) 393-4664; fax: (626) 793-3951; E-mail: zhu@hep.caltech.edu.

TABLE I  
PROPERTIES OF SOME HEAVY CRYSTAL SCINTILLATORS

Crystal	NaI(Tl)	CsI(Tl)	CsI	BaF <sub>2</sub>	BGO	PWO	LSO(Ce)	GSO(Ce)
Density (g/cm <sup>3</sup> )	3.67	4.51	4.51	4.09	7.13	8.3	7.40	6.71
Melting Point (°C)	651	621	621	1280	1050	1123	2050	1950
Radiation Length (cm)	2.59	1.85	1.85	2.06	1.12	0.9	1.14	1.37
Molière Radius (cm)	4.0	3.5	3.5	3.4	2.3	2.0	2.3	2.37
Interaction Length (cm)	41.4	37.0	37.0	29.9	21.8	18	21	22
Refractive Index <sup>a</sup>	1.85	1.79	1.95	1.50	2.15	2.2	1.82	1.85
Hygroscopicity	Yes	slight	slight	No	No	No	No	No
Luminescence <sup>b</sup> (nm)	410	560	420	300	480	560	420	440
Decay Time <sup>c</sup> (ns)	230	1300	35	6	300	50	40	60
Light Yield <sup>d,e</sup>	100	45	5.6	21	9	0.1	75	30
d(N.Y.)/dT <sup>d,f</sup> (%/°C)	~0	0.3	-0.6	-2	-1.6	-1.9	-0.3	-0.1

<sup>a</sup> At the wavelength of the emission maximum.

<sup>b</sup> Top line: slow component, bottom line: fast component.

<sup>c</sup> Relative and measured with a PMT with a Bi-alkali cathode.

<sup>d</sup> At room temperature.

calculated as [6]

$$T_e = (1 - R)^2 + R^2(1 - R)^2 + \dots = (1 - R)/(1 + R), \quad (1)$$

where

$$R = \frac{(n_{\text{crystal}} - n_{\text{air}})^2}{(n_{\text{crystal}} + n_{\text{air}})^2}. \quad (2)$$

A comparison of measured transmittance and  $T_e$  may reveal internal absorption. The excitation, photo luminescence and radiation induced phosphorescence spectra were measured by using a Hitachi F-4500 fluorescence spectrophotometer. For the excitation and emission spectra, a UV excitation light was shot to a bare surface of the sample, and the crystal was oriented so that its surface normal is at an angle  $\theta$  with respect to the excitation light. A positive  $\theta$  indicates that the photoluminescence emission light is not affected by sample's internal absorption. The scintillation light output and decay kinetics were measured by using a Photonis XP2254b PMT. A Hamamatsu R1306 with high QE was used to measure <sup>137</sup>Cs peak spectra shown in Fig. 4. A Hamamatsu RCU59 with quartz window was used to measure  $\gamma$ -ray induced anode current shown in Fig. 27. When light output was measured, one surface of the sample is coupled to the PMT with Dow Corning 200 fluid, while all other surfaces were wrapped with Tyvek paper. The light response uniformity of long samples was measured by moving a collimated  $\gamma$ -ray source along the longitudinal axis of a sample at seven points evenly distributed along the crystal and the light output response (LO) was fit to a linear function.

$$\frac{LO}{LO_{mid}} = 1 + \delta(x/x_{mid} - 1), \quad (3)$$

where  $LO_{mid}$  represents the fit result of the light output at the middle of the sample,  $\delta$  represents the deviation of the

light response uniformity, and  $x$  is the distance from the end coupled to the readout device. Because these samples have a rectangular shape, there are two ways to couple it to the PMT. We define the A end such that the sample produces a lower average light ( $LO_{mid}$ ) when it was coupled to the PMT. The other end is defined as the B end. The degradation of these optical and scintillation properties under  $\gamma$ -ray irradiation were also studied for two long LYSO samples, which were irradiated for ~22 hours each under 2, 100 and 9,000 rad/h. Fig. 2 shows  $\gamma$ -ray irradiation facilities used in this investigation.

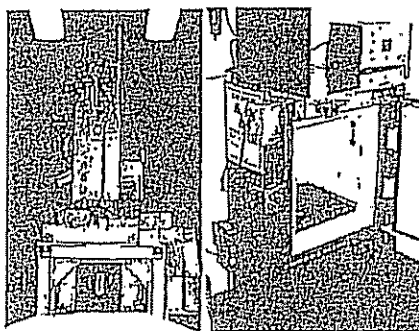


Fig. 2. A photo showing  $\gamma$ -ray irradiation facilities at Caltech. Left: an open 50 curie <sup>137</sup>Cs source provides dose rates of 2 and 100 rad/h by placing samples at different distance. Right: a closed 2,000 curie <sup>137</sup>Cs source provides a dose rate of 9,000 rad/h with 5% uniformity.

SMS002418

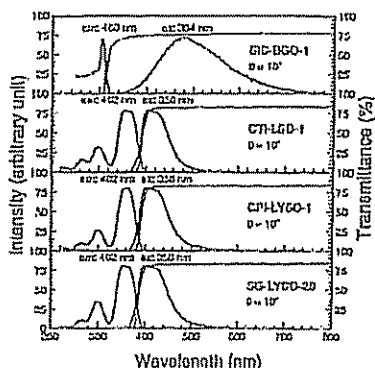


Fig. 3. Optical properties as a function of wavelength for four 1.7 cm cubic samples. The emission (red) and excitation (blue) spectra correspond to the left vertical scale, and the transmittance (green) spectra to the right. The  $\theta$  refers to the angle between crystal normal and the incident excitation light direction as discussed in text.

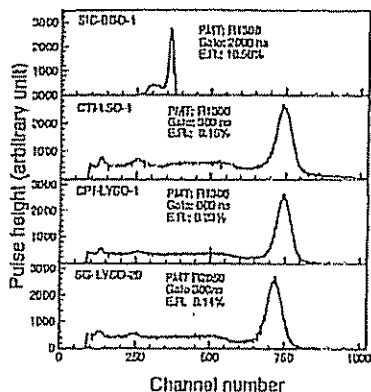


Fig. 4.  $^{137}\text{Cs}$  peak spectrum measured with the R1306 PMT is shown for four 1.7 cm cubic samples.

## II. SCINTILLATION AND OPTICAL PROPERTIES

Fig. 3 shows a comparison of transmittance (right scale), emission and excitation spectra for four 1.7 cm cubic samples: a SIC BGO, a CTI LSO and two LYSO samples from CPI and Saint-Gobain. One notes that the LSO and LYSO samples have identical transmittance, excitation and emission spectra. It is also interesting to note that the emission spectra of LSO and LYSO are overlapping with their absorption edge

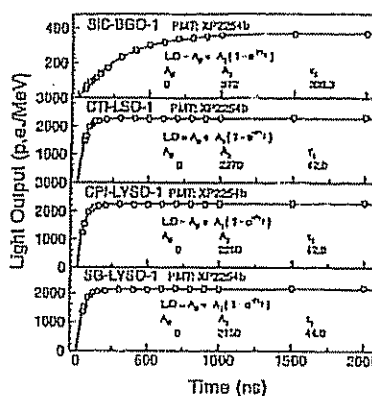


Fig. 5. Light output measured with the XP2254b PMT is shown as a function of integration time for four 1.7 cm cubic samples.

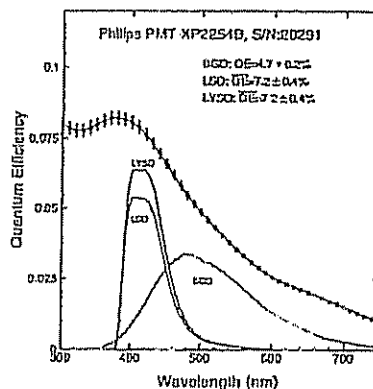


Fig. 6. The scintillation efficiency of the XP2254b PMT is shown as function of wavelength together with emission spectra of BGO, LSO and LYSO samples.

in transmittance, while the emission of BGO is well within its transparent wavelength region. This indicates that the light output of LSO and LYSO is strongly affected by their intrinsic absorption edge. Alternatively, a push of transmittance edge to short wavelength would effectively increase its light output for LSO and LYSO crystals. Fig. 4 shows a comparison of  $^{137}\text{Cs}$  peak observed by 1.7 cm cubic samples coupled in the R1306 PMT. The FWHM resolution of the  $^{137}\text{Cs}$  peak observed is 8 to 9% for LSO and LYSO and 10% for BGO. Fig. 5



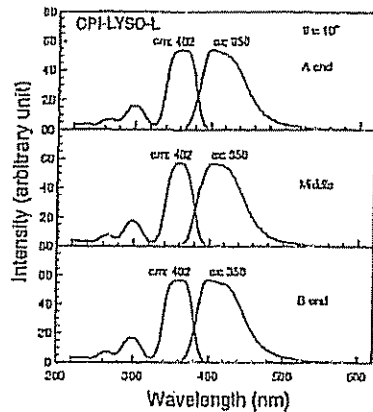


Fig. 7. Excitation (red) and emission (blue) spectra measured for the CPI long LYSO sample at three locations.

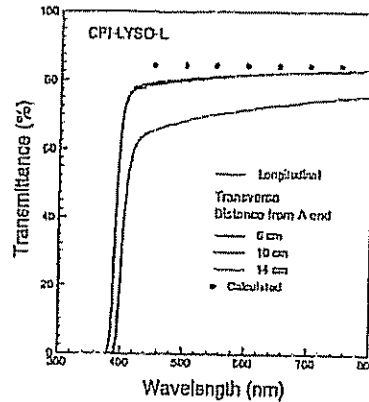


Fig. 9. Longitudinal (black) and transverse (color) transmittance spectra are shown as a function of wavelength for the CPI long LYSO sample, and compared to the calculated threshold level.

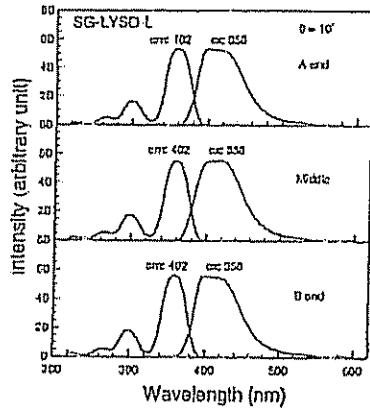


Fig. 8. Excitation (red) and emission (blue) spectra measured for the Saint-Gobain long LYSO sample at three locations.

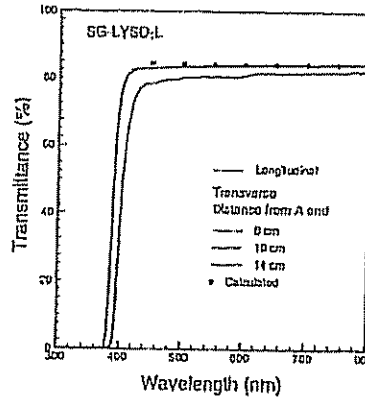


Fig. 10. Longitudinal (black) and transverse (color) transmittance spectra are shown as a function of wavelength for the Saint-Gobain long LYSO sample, and compared to the calculated threshold level.

shows a comparison of decay kinetics observed by 7.7 cm cubic samples coupled to the XP2254b PMT. One notes that the LSO and LYSO samples have consistent decay time and photoelectron yield. While the 300 ns decay time of BGO is about a factor of 7 slower, its measured photoelectron yield is also about a factor of 7 lower than that of LSO and LYSO. Fig. 6 shows the quantum efficiency of the XP2254b PMT used to measure the

light output and decay kinetics. Also shown in the figure are the emission spectra and emission weighted average quantum efficiencies for these samples, which may be used to convert measured photoelectron yield to an absolute light out in photon numbers. Taking into account the PMT response, we conclude that the amplitude of BGO light output is about a factor of 4 lower than that of LSO and LYSO.

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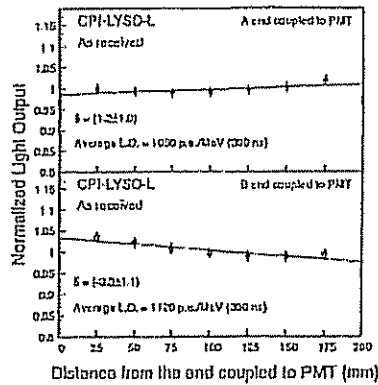


Fig. 11. Light response uniformity of the CPI long LYSO sample measured with the A (top) and B (bottom) end coupled to the XP2254b PMT.

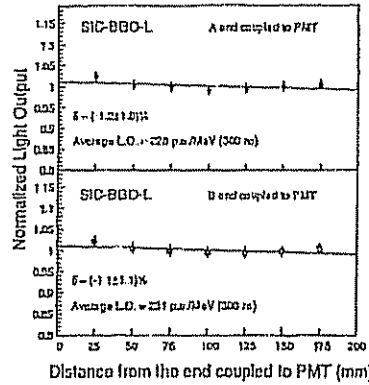


Fig. 13. Light response uniformity of the SIC long BGO sample measured with the A (top) and B (bottom) end coupled to the XP2254b PMT.

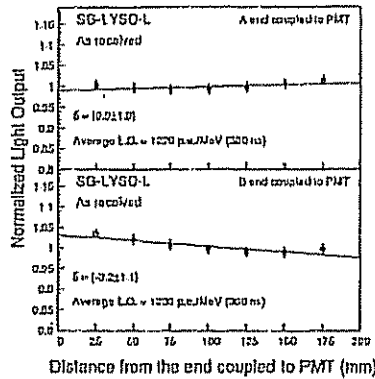


Fig. 17. Light response uniformity of the Saint-Gobain long LYSO sample measured with the A (top) and B (bottom) end coupled to the XP2254b PMT.

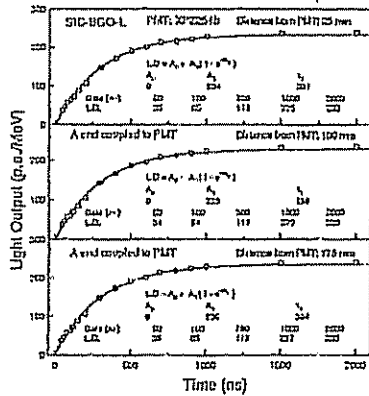


Fig. 14. The decay kinetics of the SIC long BGO sample measured at three locations with the A end coupled to the XP2254b PMT.

Fig. 7 and 8 show excitation and emission spectra measured for two long LYSO samples. No variation is observed in spectra measured at three locations along these two crystals, indicating a good longitudinal uniformity. Fig. 9 and 10 show longitudinal and transverse transmittance spectra measured for two long LYSO samples. Once again, no variation is observed in transverse transmittance spectra measured at three locations along these two crystals. While the transverse transmittance measured for the Saint-Gobain sample approaches the theoretical limit calculated according to Equation 1, its longitudinal

transmittance shows an absorption peak at 580 nm, which does not interfere with its emission so has no effect on its light output. A poorer transmittance was observed for the CPI sample, which may be attributed to its poor surface polishing.

Fig. 11, 12 and 13 show light response uniformity measured by using the XP2254b PMT and the corresponding linear fit to the equation 3 for three long samples. While the BGO sample shows a consistent slight negative  $\delta$  for both the A and B end coupled to the PMT, both LYSO samples show different

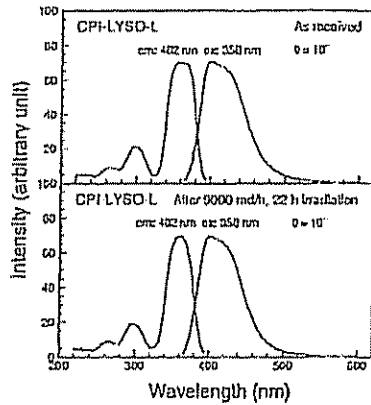


Fig. 15. Excitation (red) and emission (blue) spectra measured for the CPI long LYSO sample before and after 9,000 rad/h irradiation.

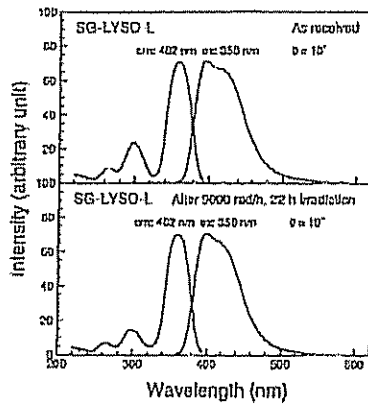


Fig. 16. Excitation (red) and emission (blue) spectra measured for the Saint-Gobain long LYSO sample before and after 9,000 rad/h irradiation.

sign of  $\delta$  when the end coupled to the PMT is changed. This observation hints a slight longitudinal non-uniformity of light yield along the axis of long LYSO samples. The BGO sample, on the other hand, has a good longitudinal uniformity as shown in Fig. 14. This non-uniformity in LYSO may be attributed to the cerium concentration [7] or, less likely, to the fraction of yttrium in LYSO [8].

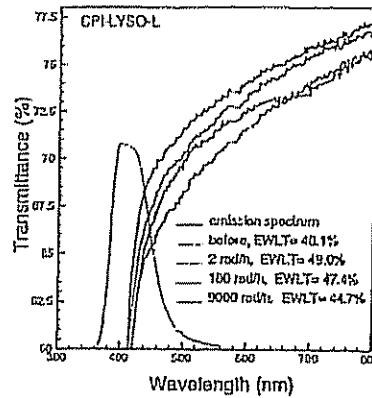


Fig. 17. Longitudinal transmittance spectra before and after 2, 100 and 9,000 rad/h irradiation and the emission spectrum are shown as a function of wavelength for the CPI long LYSO sample.

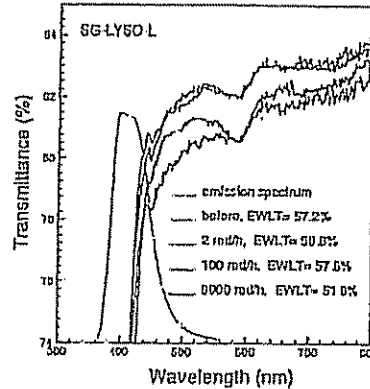


Fig. 18. Longitudinal transmittance spectra before and after 2, 100 and 9,000 rad/h irradiation and the emission spectrum are shown as a function of wavelength for the Saint-Gobain long LYSO sample.

### III. RADIATION HARDNESS

All known crystal scintillators suffer from radiation damage. Crystal radiation damage appears in radiation induced absorption (color center formation), reduced scintillation light yield (damage of the scintillation mechanism) or radiation induced phosphorescence (afterglow). While the damage to the absorption and scintillation would lead to a reduction of

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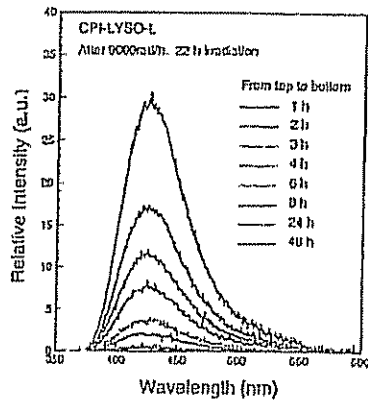


Fig. 21. Radiation induced phosphorescence spectra are shown as a function of wavelength for the CPI long LYSO sample after 9,000 rad/h irradiation.

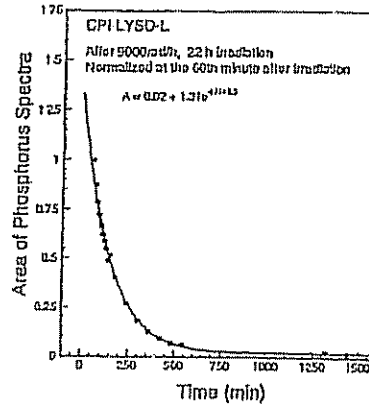


Fig. 25. The amplitude of phosphorescence is shown as a function of time after 9,000 rad/h irradiation for the CPI long LYSO sample.

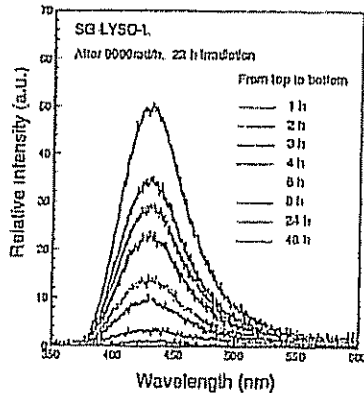


Fig. 24. Radiation induced phosphorescence spectra are shown as a function of wavelength for the Saint-Gobain long LYSO sample after 9,000 rad/h irradiation.

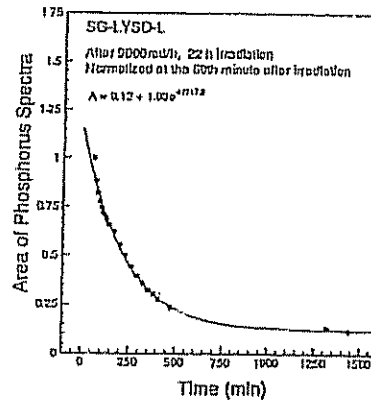


Fig. 26. The amplitude of phosphorescence is shown as a function of time after 9,000 rad/h irradiation for the Saint-Gobain long LYSO sample.

after 9,000 rad/h irradiation and the corresponding linear fit to the equation 3 for CPI and Saint-Gobain LYSO samples. Compared to Figs. 11 and 12, there are slight decrease of the slope ( $\delta$ ) as well as average light output ( $LO_{avg}$ ) after irradiation, indicating an increase of internal absorption. Fig. 21 and 22 compare the decay kinetics before and after 9,000 rad/h irradiation. While some degradation of light output is observed, the decay time remains stable for both samples.

Fig. 23 and 24 show radiation induced phosphorescence spectra measured after 9,000 rad/h irradiation for CPI and Saint-Gobain LYSO samples. Similar phosphorescence spectrum peaked at 430 nm was observed for both samples. The amplitude of phosphorescence, normalized to 1 h after the end of irradiation, were fit to an exponential function

$$A = A_0 + A_1 e^{-t/\tau} \quad (4)$$

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TABLE II  
 $\gamma$ -RAY INDUCED READOUT NOISE IN LARGE SIZE LYSO SAMPLES

Sample ID	L.Y. p.e./MeV	F $\mu A rad^{-1}h$	$Q_{10}$ p.e.	$Q_{500}$ p.e.	$\sigma_{10}$ MeV	$\sigma_{500}$ MeV
CPI	1.480	41	$6.98 \times 10^4$	$2.33 \times 10^5$	0.18	1.03
Saint-Gobain	1.580	42	$7.15 \times 10^4$	$2.38 \times 10^5$	0.17	0.97

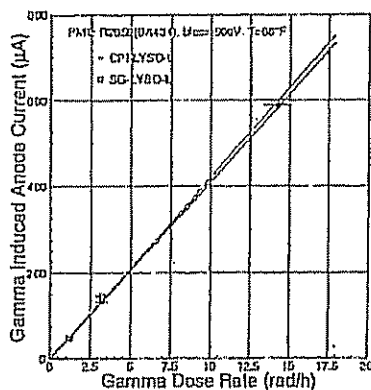


Fig. 27. The  $\gamma$ -ray induced anode current is shown as a function of the dose rate for the CPI and Saint-Gobain large LYSO samples.

The result of the fit is shown in Fig. 25 and 26. The decay time of radiation induced phosphorescence is determined to be 2.5 to 3 h, and the Saint-Gobain sample is noticed to have a little larger residual phosphorescence.

To evaluate the radiation induced phosphorescence related readout noise, the Hamamatsu R2059 PMT was used to measure the  $\gamma$ -ray induced anode current for LYSO samples under  $\gamma$ -ray irradiations at 2, 100 and 9,000 rad/h. Fig. 27 shows the result of this measurement and the corresponding linear fit. These two samples show compatible radiation induced phosphorescence. Table III summarizes the result, where L.Y. is the photoelectron yield of the sample as measured by the R2059 PMT. F is the  $\gamma$ -ray induced anode current per unit dose rate from the fit.  $Q_{10}$  and  $Q_{500}$  are the induced photoelectron numbers in 100 ns gate for these samples under 15 and 500 rad/h respectively, which were calculated by using F and the corresponding gain of R2059 with 900 V bias.  $\sigma_{10}$  and  $\sigma_{500}$  are the corresponding energy equivalent readout noise, which were derived as the eq. 2. Fluctuation of the photoelectron numbers. The radiation induced phosphorescence related readout noise is estimated to be about 1 MeV equivalent with 100 ns integration time under 500 rad/h.

#### IV. SUMMARY

Ce doped LSO and LYSO crystals have identical emission, excitation and transmittance spectra. The amplitude of their fast scintillation light output of 42 ns decay time is about 4 times of BGO with 300 ns decay time. The absorption edge in their transmittance spectrum affects their light output. One approach to increase their light output is to move their absorption edge to a shorter wavelength. Large size ( $2.5 \times 2.5 \times 20$  cm) LYSO samples from CPI and Saint-Gobain have good overall longitudinal uniformity in optical and scintillation properties. Their light response uniformly, however, may slightly be affected by the distribution of the Ce concentration.

Radiation effect on transmittance, emission and light output in LYSO samples in an environment of below 100 rad/h is small as compared to other commonly used crystals. Radiation induced phosphorescence in LYSO has a time constant of 2.5 to 3 h. The  $\gamma$ -ray induced phosphorescence related readout noise is about 1 MeV equivalent calculated in 100 ns integration time for  $2.5 \times 2.5 \times 20$  cm LYSO samples in an radiation environment of 500 rad/h.

In a brief summary, with existing mass production capabilities, LSO and LYSO crystals are a good candidate for applications in high energy and nuclear physics. Further investigation, however, is needed to understand the consequence of radiation damage for large size samples readout with solid state devices, such as Si PD or APD.

#### ACKNOWLEDGMENTS

Useful discussions with Drs. C. Melcher and B. Choi are acknowledged.

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**EXHIBIT 13**

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